Deceleration and Trapping of Cold Free Radicals by Pulsed Magnetic Fields

Towards Ultra-high Resolution Spectroscopy and Cold Chemistry

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Cold Molecules

Molecules with a low translational energy

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1. Ultra-high Resolution/Precision Spectroscopy

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The Origin of spectral linewidth

Natural $$\frac{1}{2\pi\tau_{sp}}$$

Doppler $$\sqrt{\pi \ln 2}\ \tilde{\nu}\langle v\rangle$$

Transit time $$\frac{1}{2\sqrt{2\ln 2}}\frac{\langle v\rangle}{d}$$

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CH_3 (M = 15) ν_3 (3,160 cm^{-1}) μ_{10}=0.1 D

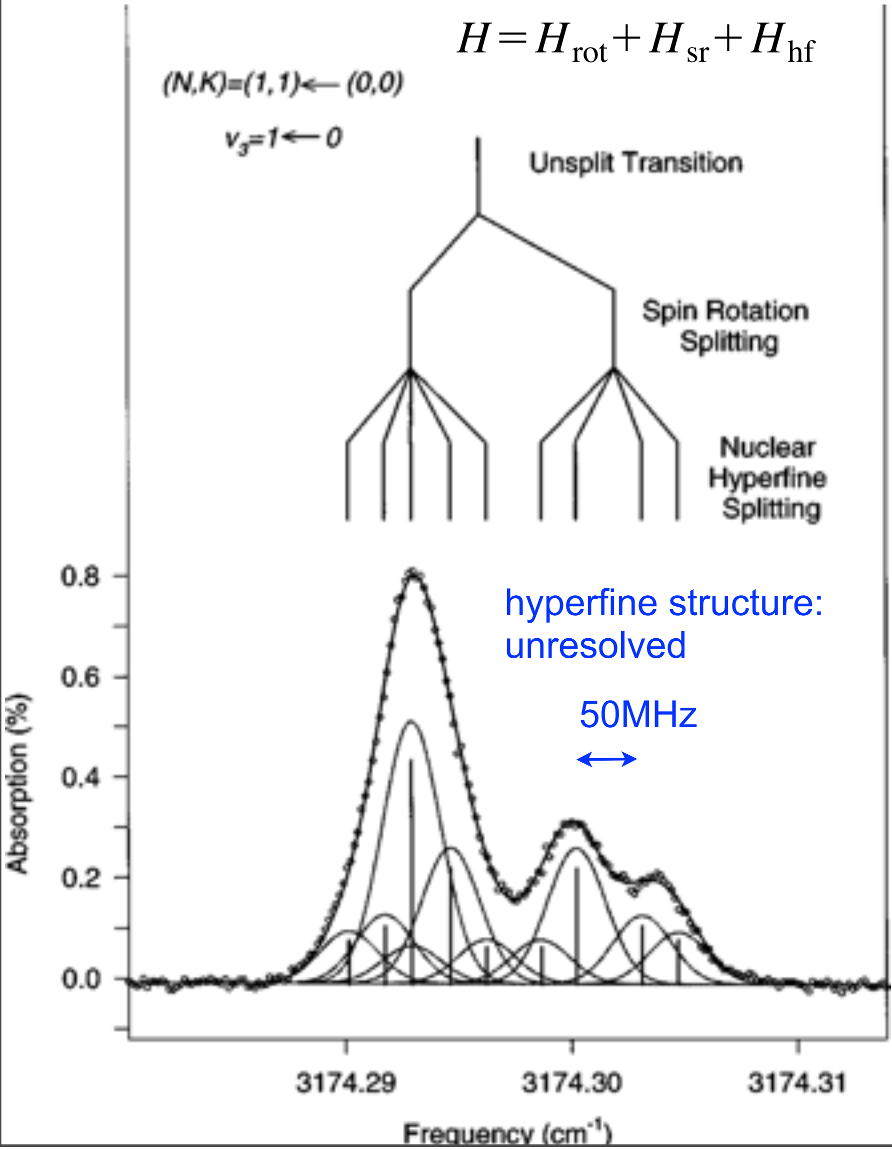
T	Mean velocity <v> [m/s]	Doppler [MHz]	Transit time* [kHz]	Natural [Hz]
300 K	650	303	276	16
1 K	38	17.5	16	16
1 mK	1.2	0.55	0.5	16

***1 mm diameter laser radiation**

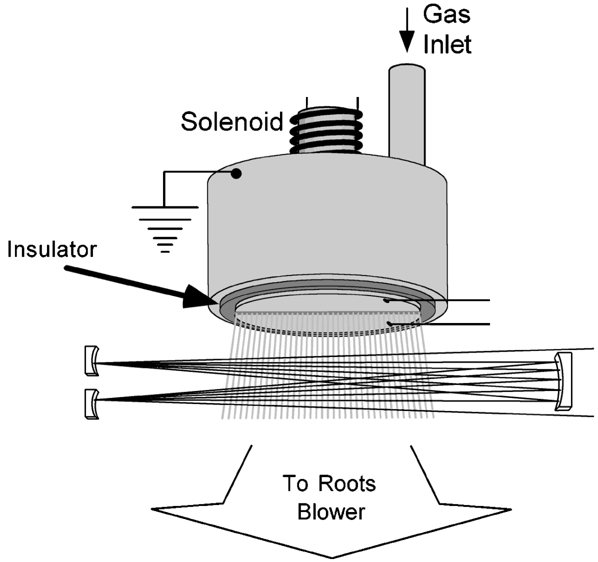
High-Resolution Spectroscopy

ν_3 Absorption of CH_3 (slit jet)

Davis et al. J. Chem. Phys. 107, 5661 (1997)



solid line. The least-squares fits indicate an OH Doppler width of $\Delta\nu = 105(2)$ MHz, which is in reasonable agreement with $\Delta\nu = 98(3)$ MHz predictions from scaling the experimentally observed HF dimer Doppler widths by the ratio of absorption frequencies and the square root of the mass. This would be consistent with a slit translational temperature of $\approx$30 K and $\approx$fourfold enhancement in spectral resolution over Doppler limited linewidths under typical discharge cell conditions.



30 K ~ 200 m s^{-1}

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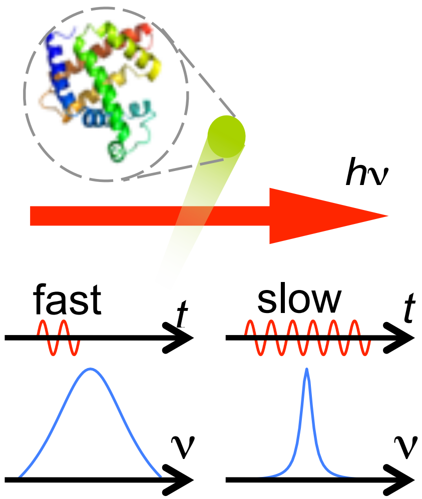
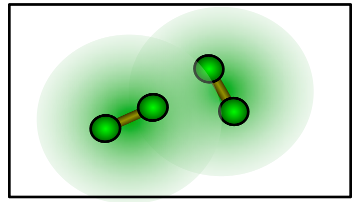


Ultrahigh-resolution spectroscopy
Test of fundamental symmetries
Constancy of fundamental constants
eEDM measurement

Cold Molecules

2. Collision Dynamics / Coherent Control



Relevant length: Thermal de Broglie length

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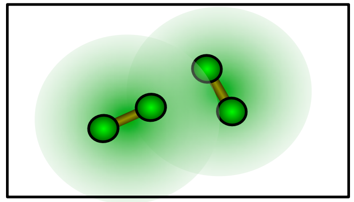

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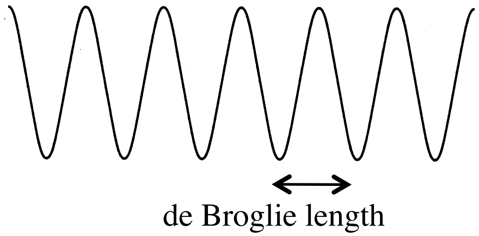
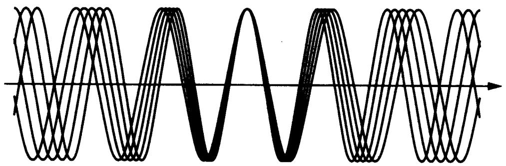
matter wave



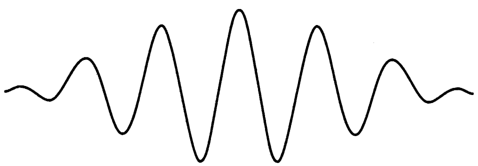
$$\lambda_{\mathrm{dB}} = \frac{h}{p}$$

thermal fluctuation of momentum



$$\Delta p \sim \sqrt{3mk_{\mathrm{B}}T}$$

superposition of matter wave



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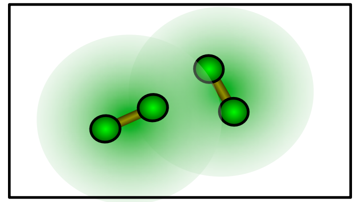

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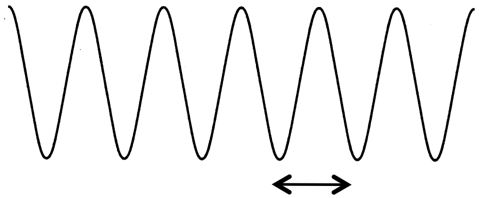

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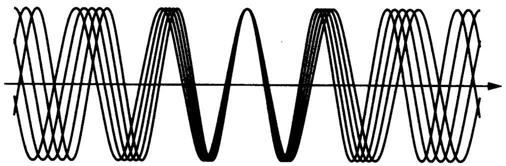

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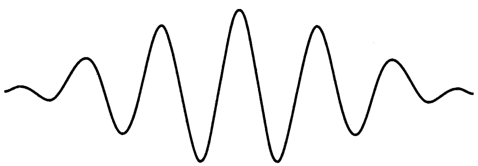

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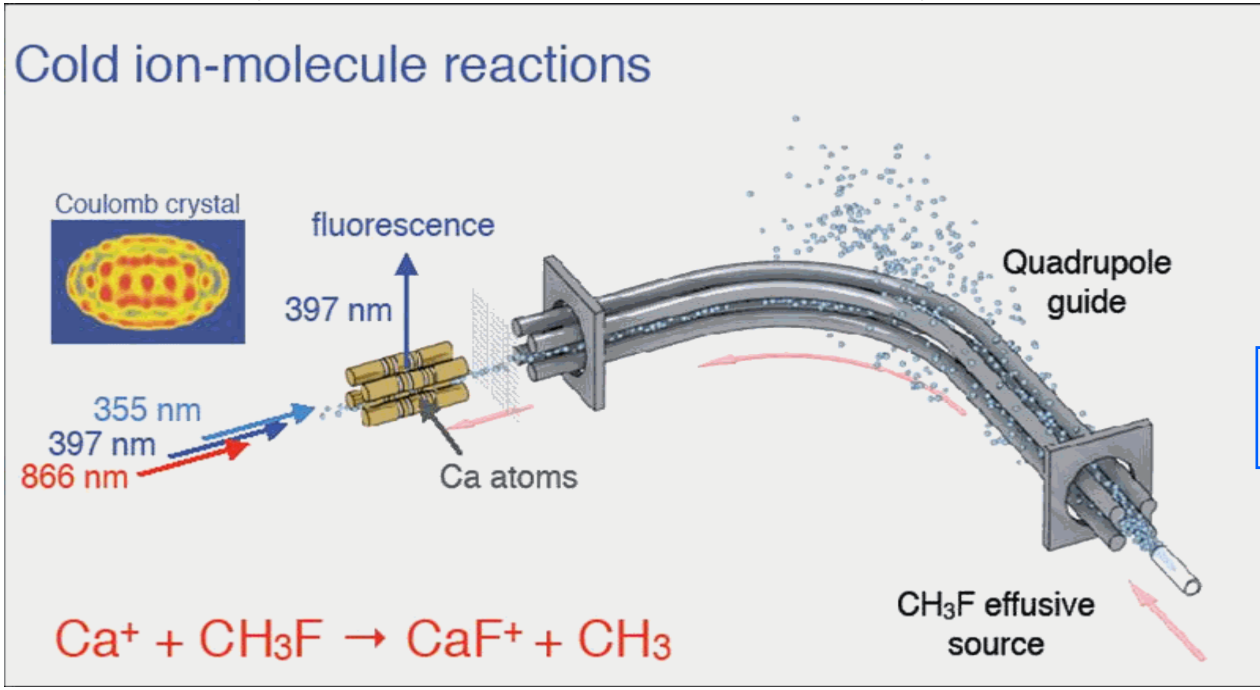
thermal de Broglie length

	H_2	CH_4	C_6H_6
M(amu)	2	16	78
300K	70 pm	25 pm	11 pm
1 K	1.3 nm	440 pm	200 pm
0.1 mK	130 nm	44 nm	20 nm
10 nK	13 μm	4.4 μm	2 μm
“classical” size	1Å 100 pm	3Å 300 pm	5Å 500 pm

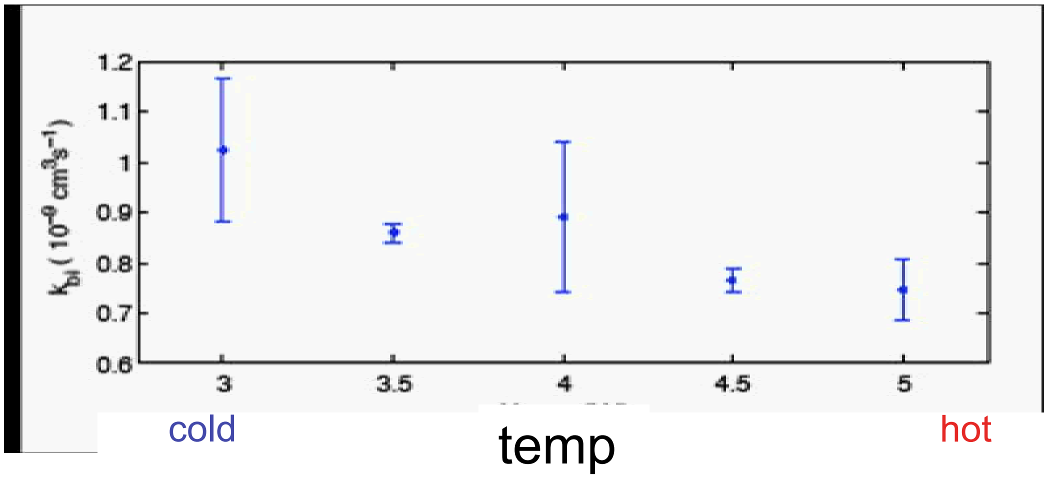
large matter wave : below 1 K

Cold Ion-Molecule Reactions

Tim Softley's Group http://physchem.ox.ac.uk/~tps/iquad.htm



Negative temperature effect in ion-neutral reaction



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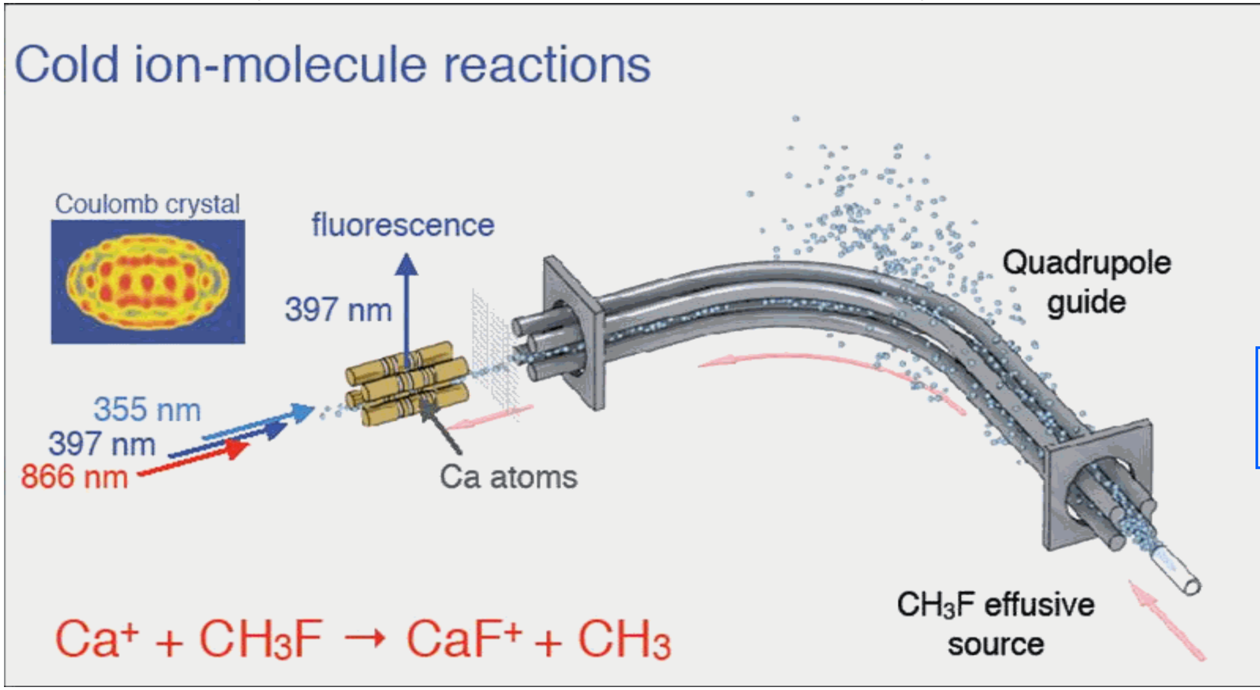
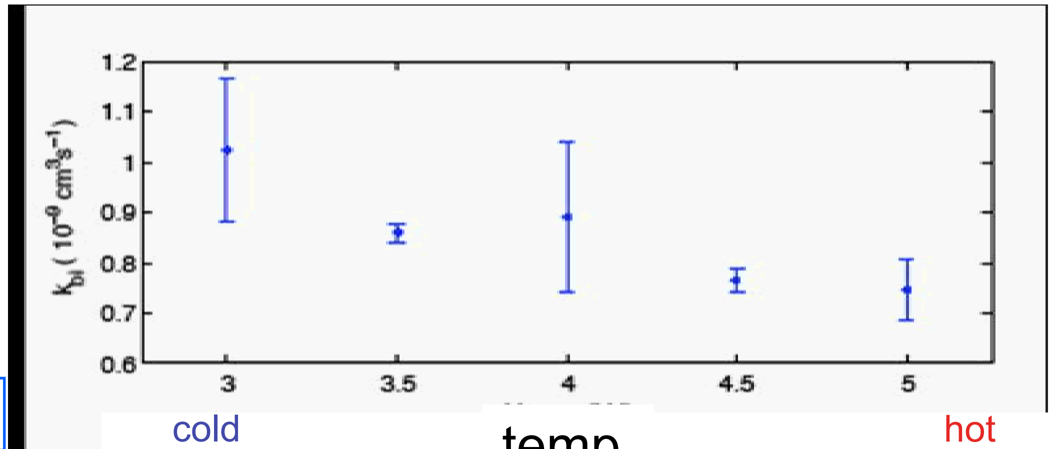


Negative temperature effect in ion-neutral reaction

Interstellar Chemistry

T: 5K - 100 K

neutral-neutral reactions?



cold temp hot

Cold Molecules

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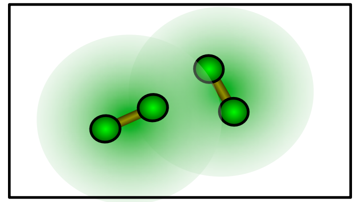

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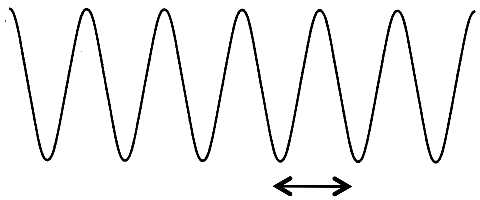

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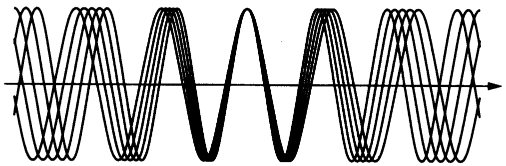

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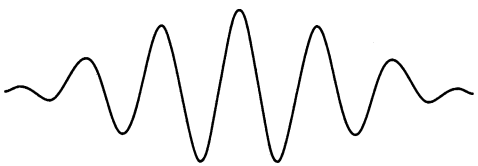

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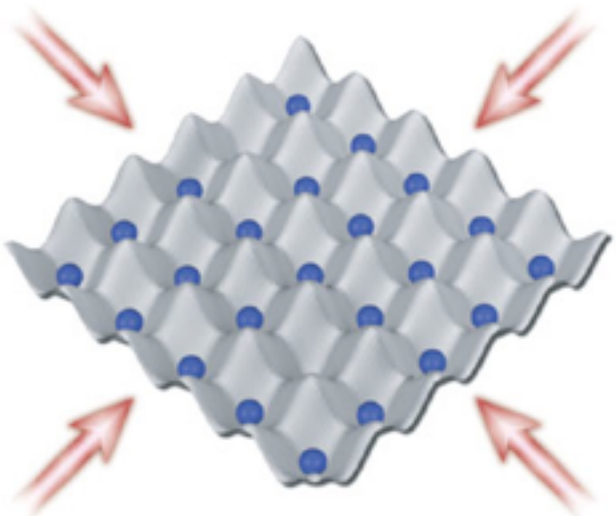
Collision and reaction dynamics
Interference of molecular dB waves
Coherent control of reactions
Separation of chiral molecules
Quantum information processes

Motivation

Spatially confine (chemically unstable) molecules

Ultra-high-resolution spectroscopy
Cold Chemistry



DC static fields
- **Paul trap**
- **Anti-Helmholtz trap**

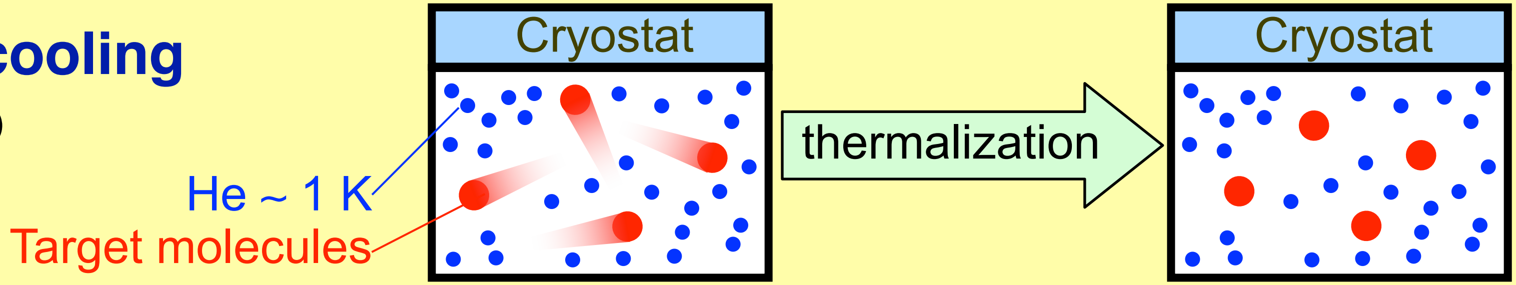
AC fields
- **Optical lattice**
- **MW trap**

translational velocity : $v<10\ m\ s^{-1}$

Generation of Translationally Cold Molecules

Buffer gas cooling

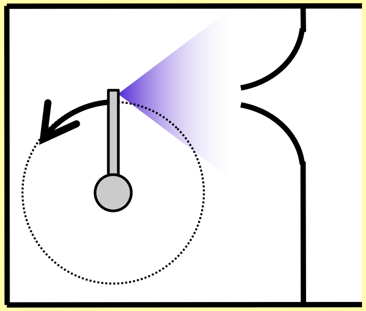
Doyle (Harvard)



Counter rotating source

- Mechanical deceleration

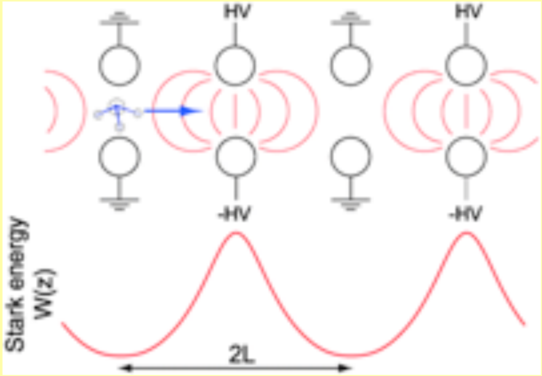
Herschbach (Harvard)
Stienkemeier, Mudrich (Freiburg)
Momose (UBC)

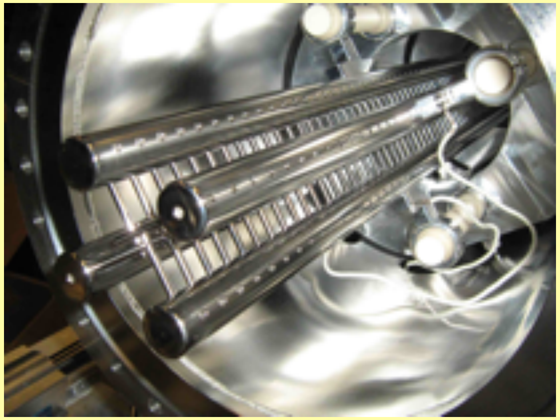


Stark effect on polar molecules

- Stark deceleration

Meijer (Max-Planck Institute) etc





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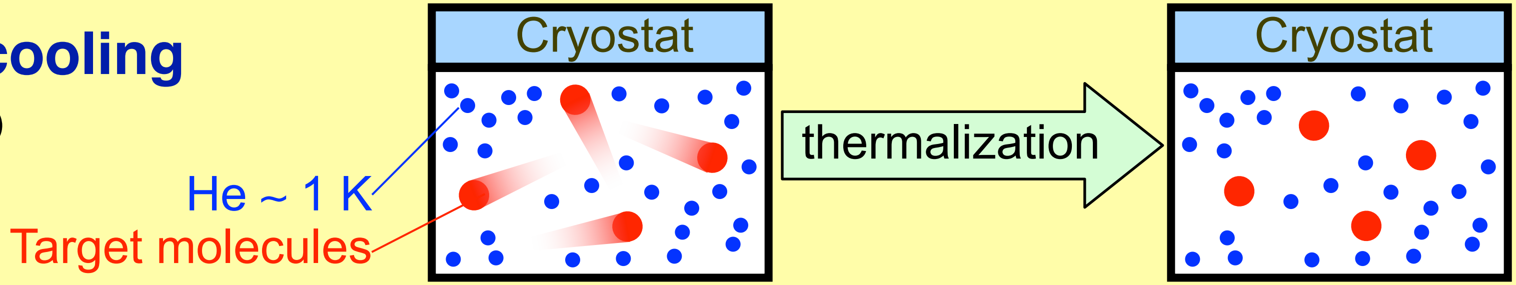

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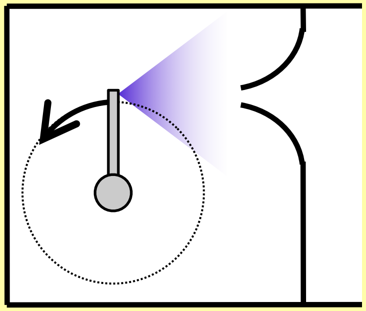

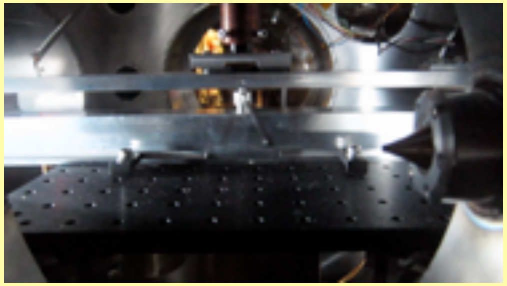

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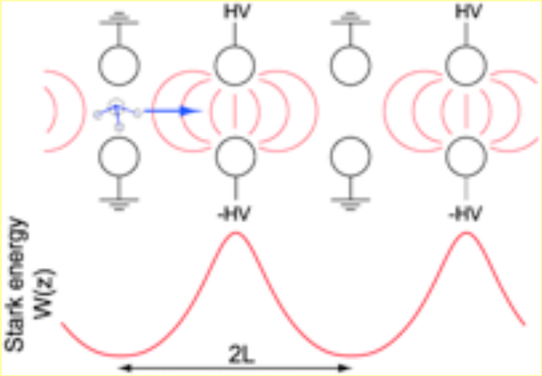
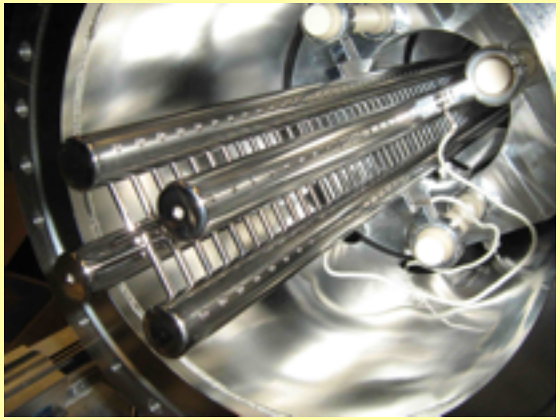
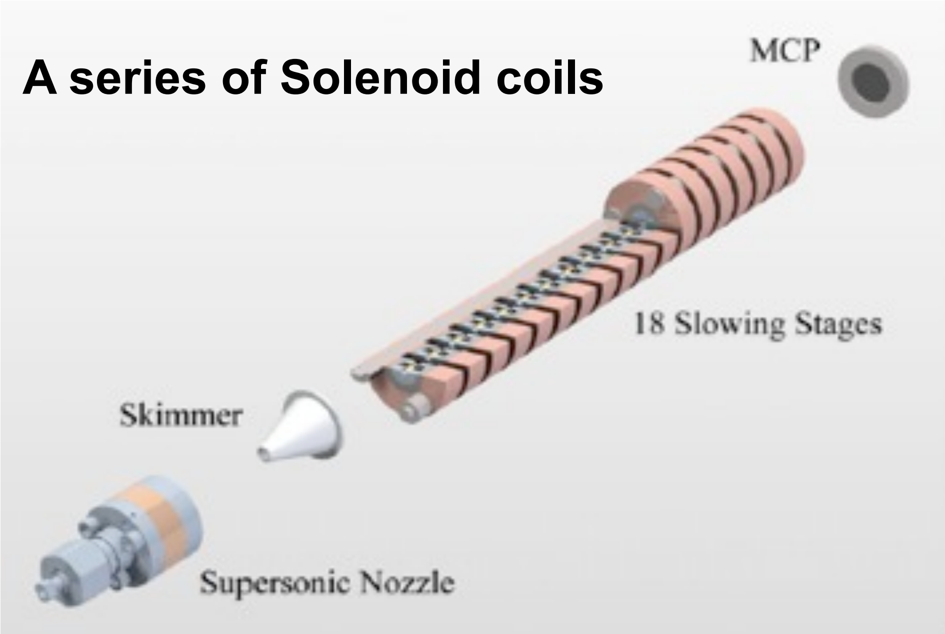




Zeeman Decelerator



H atoms F. Merkt et al. PRA **76**, 023412 (2007).
Ne atoms MG Raizen, PRL **100**, 090303 (2008).

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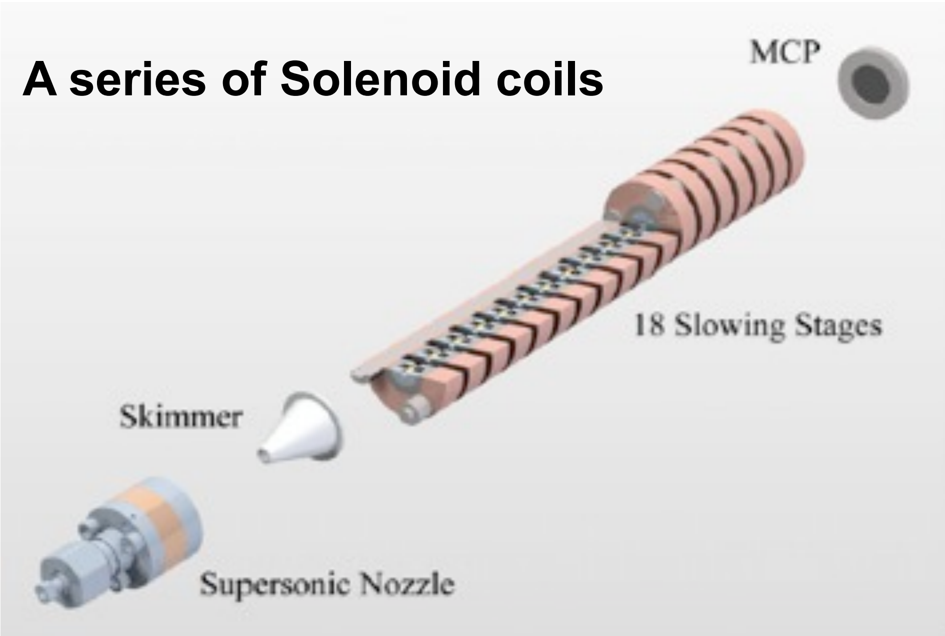
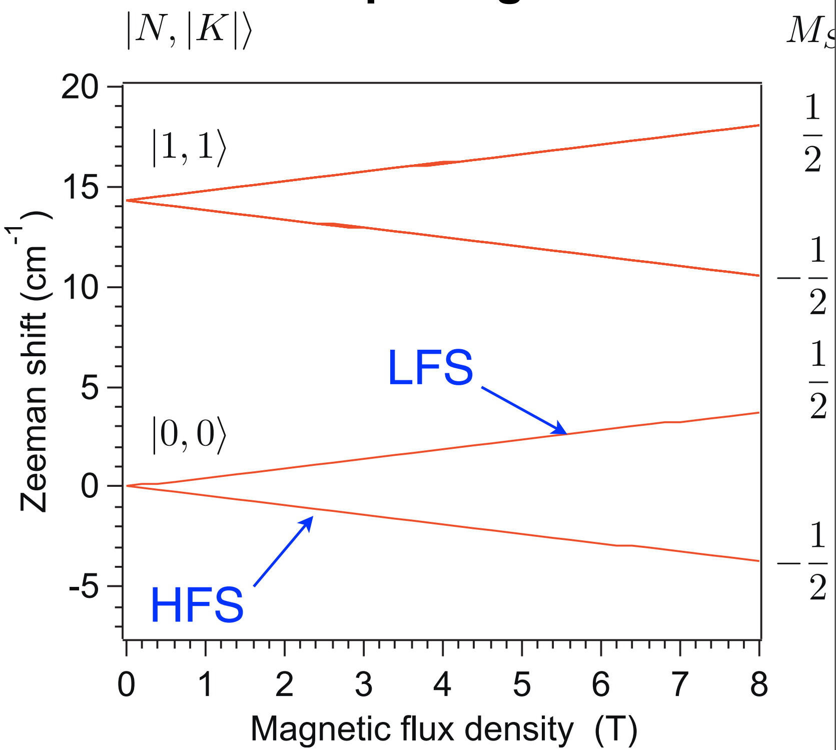


Zeeman splitting of CH_3



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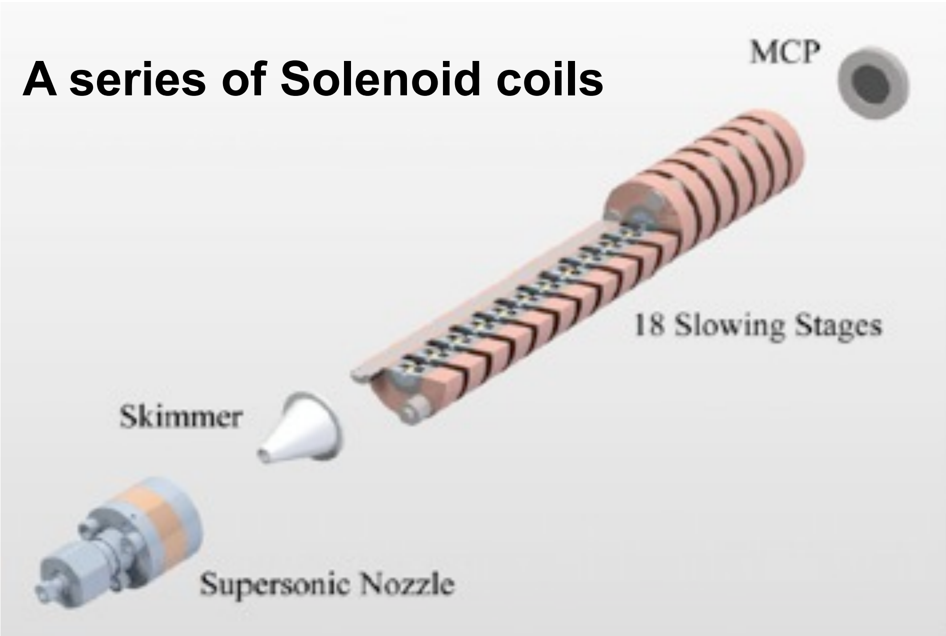

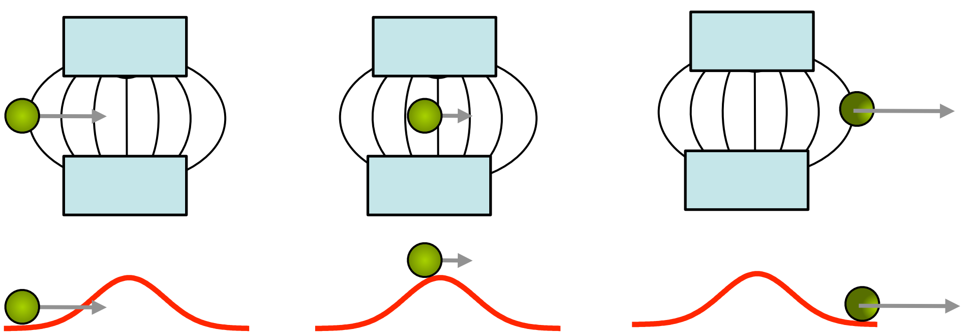

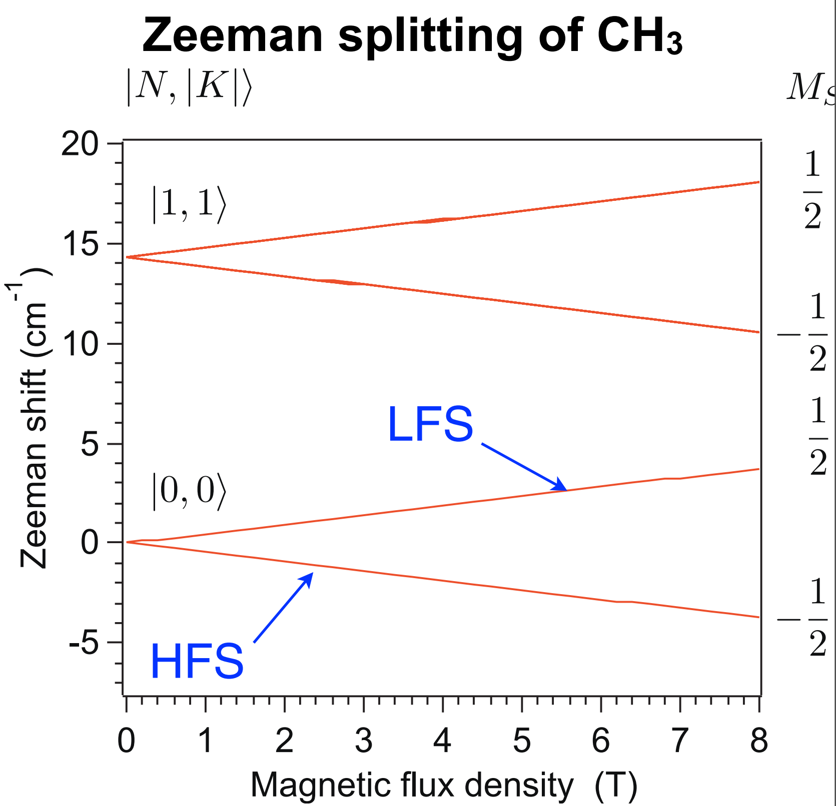

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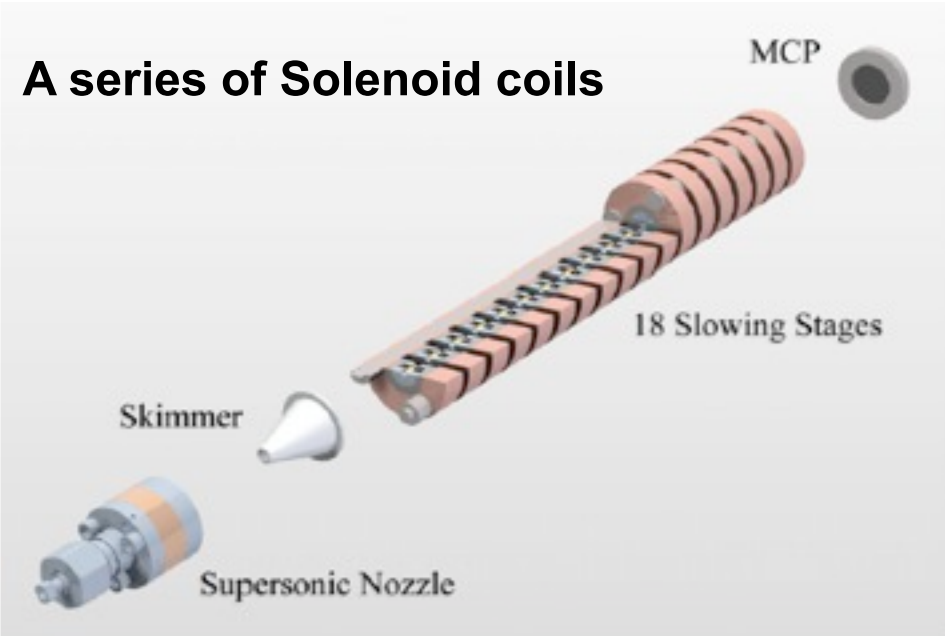

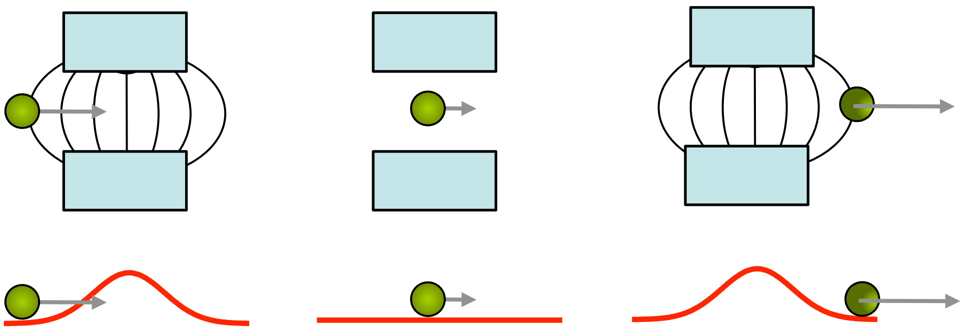

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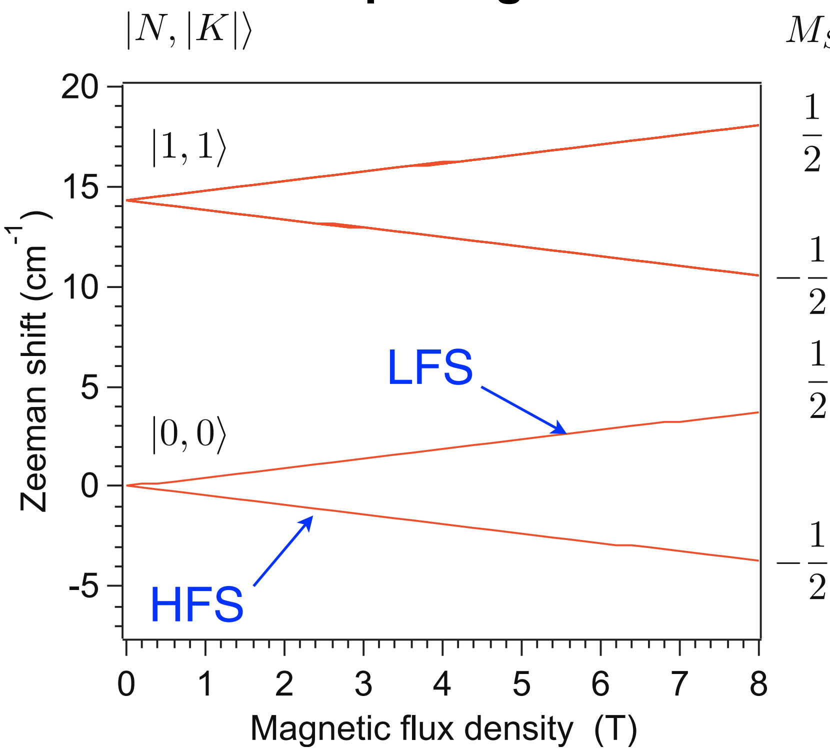

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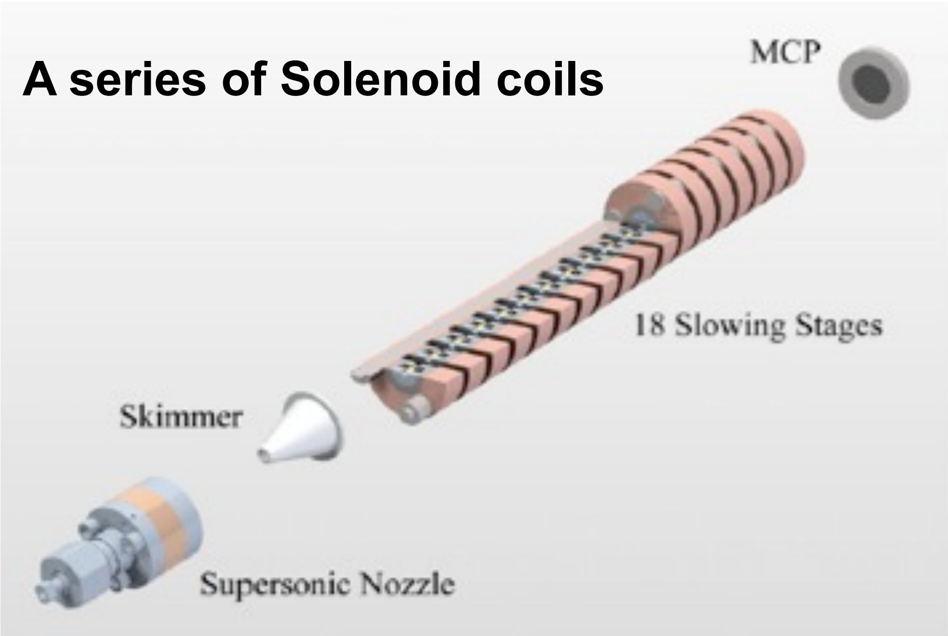

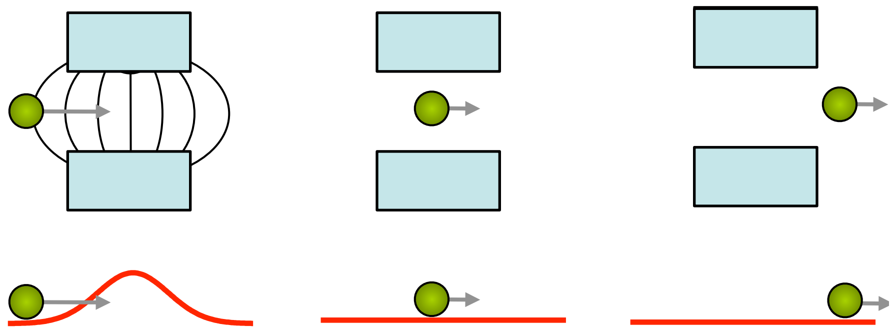

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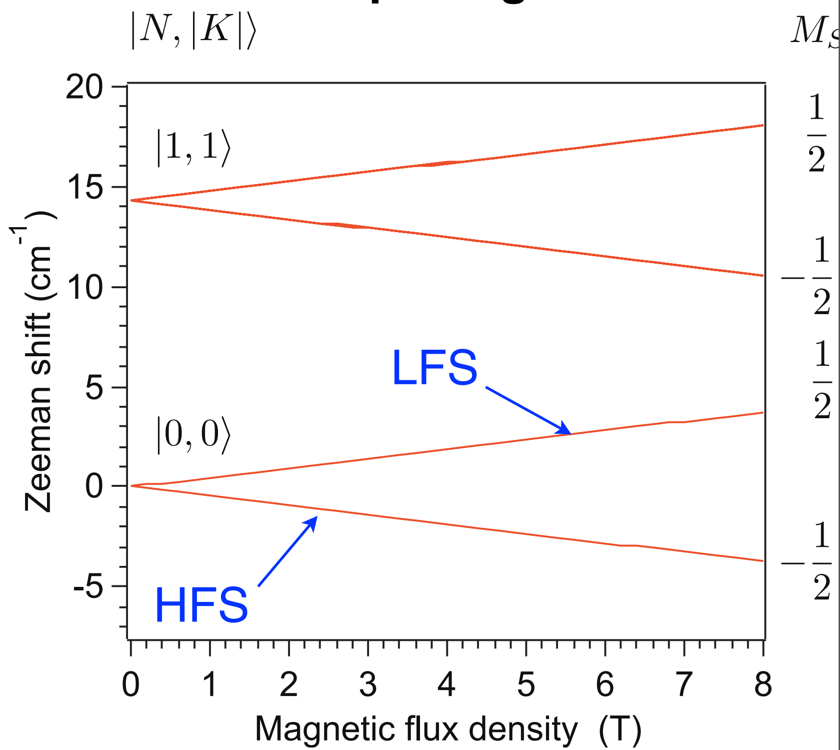

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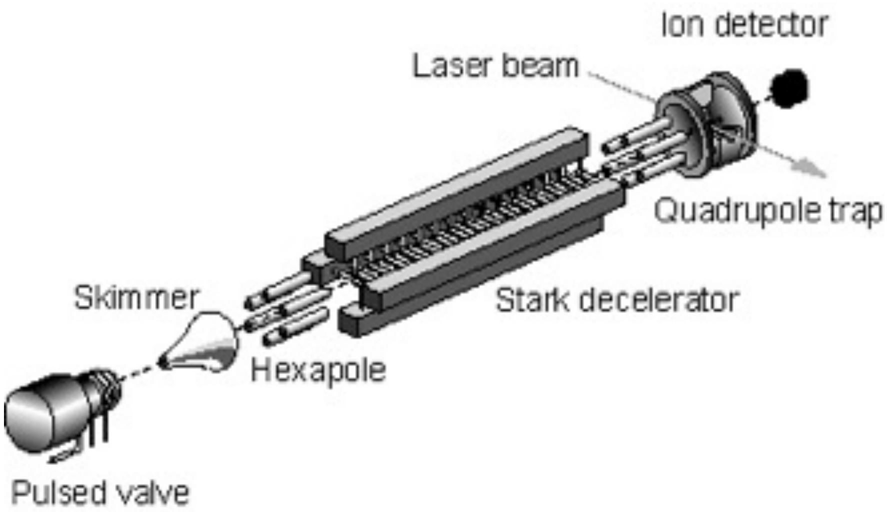
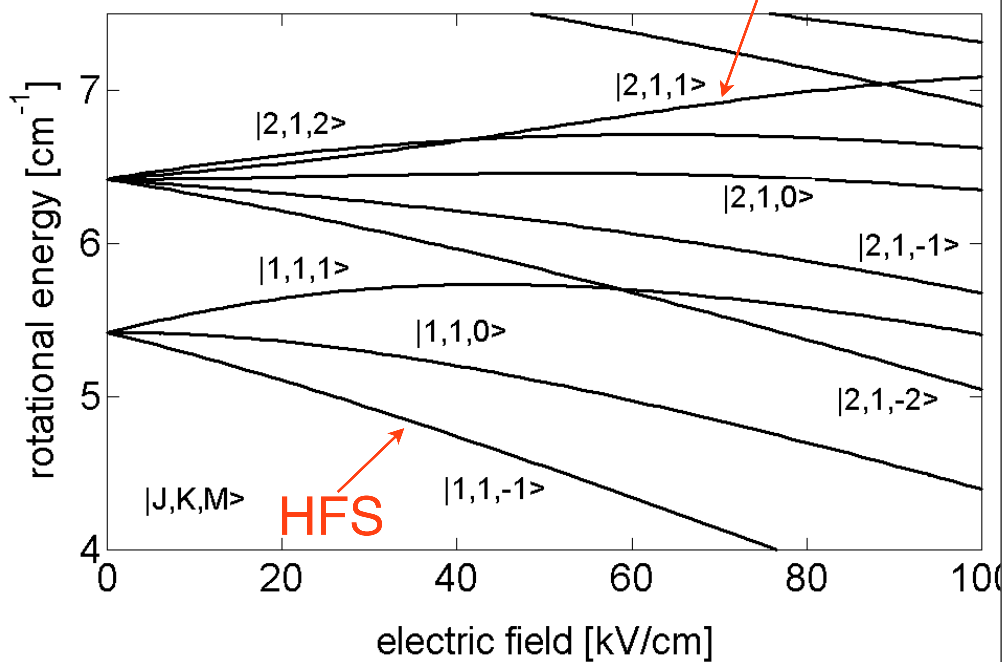
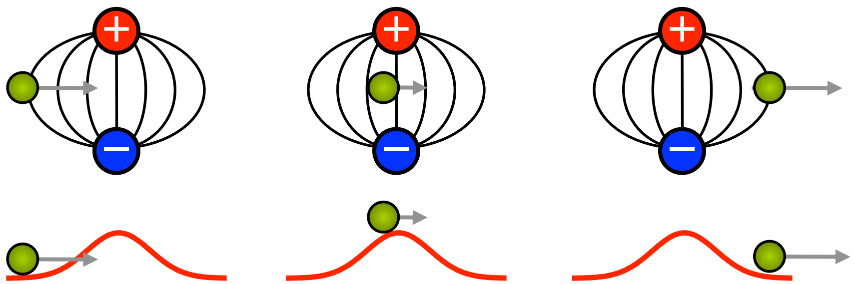
Stark Decelerator



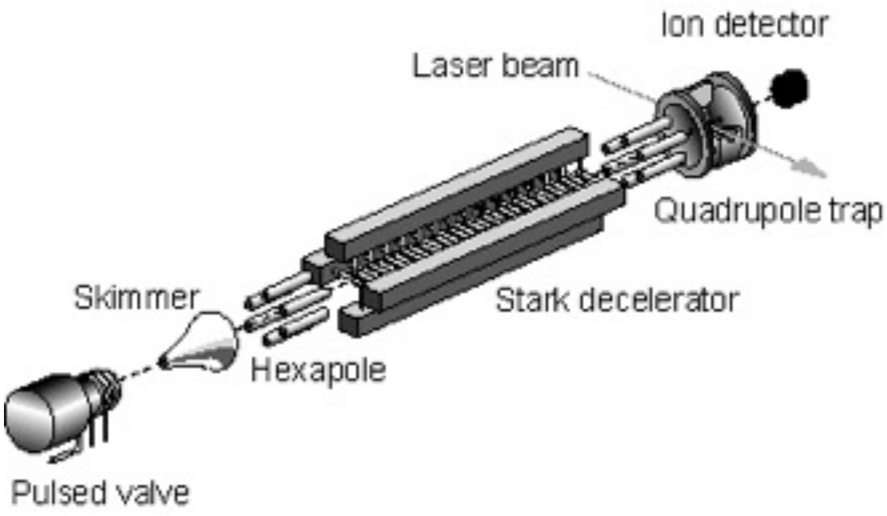
Stark splitting of CH_3I

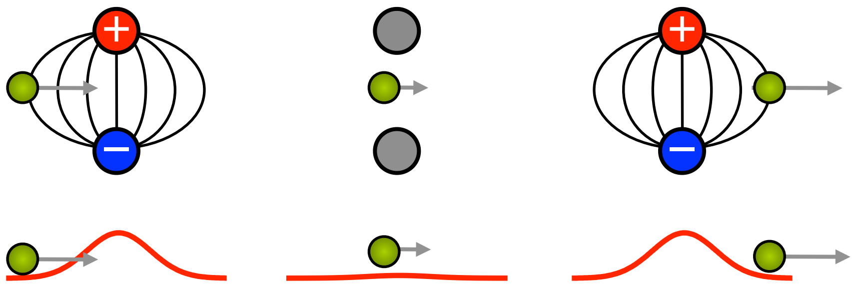




Meijer et al,
Phys. Rev. A **65**, 53416 (2002)

Stark Decelerator





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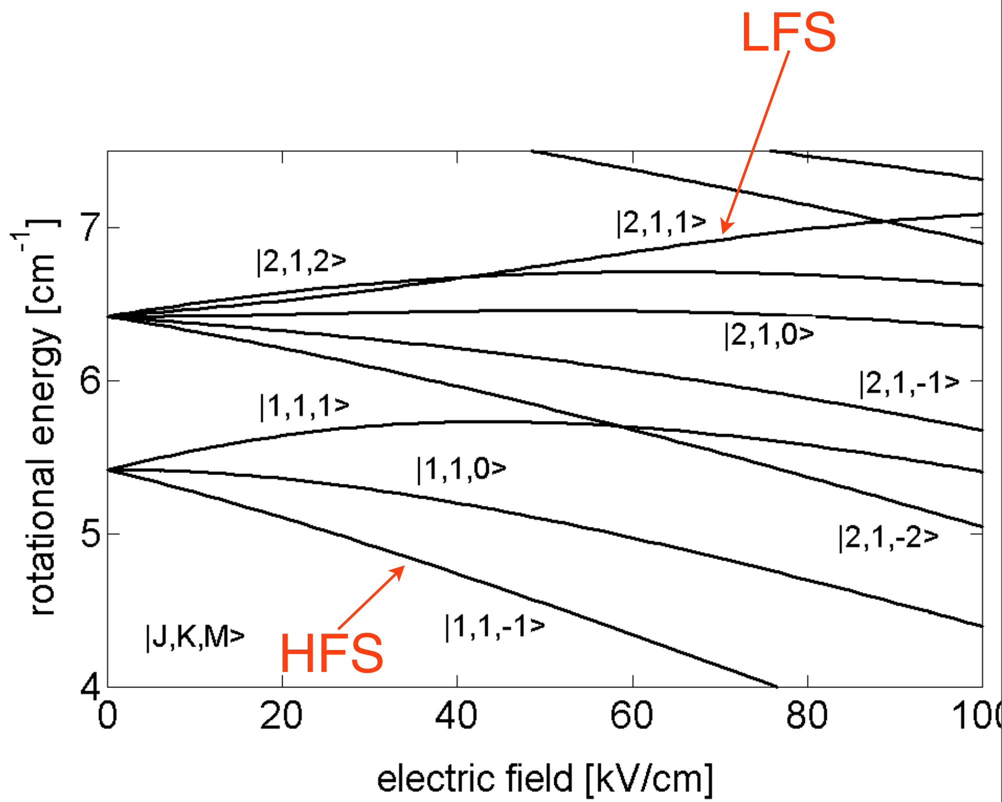

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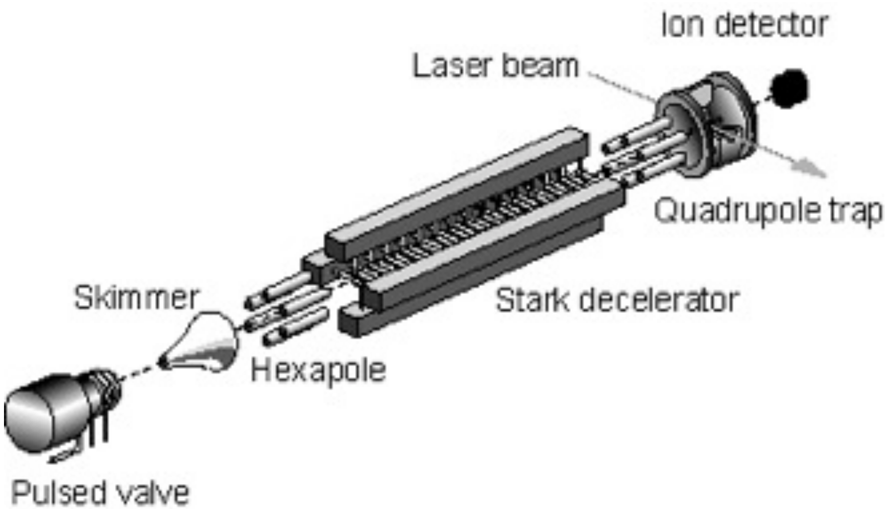

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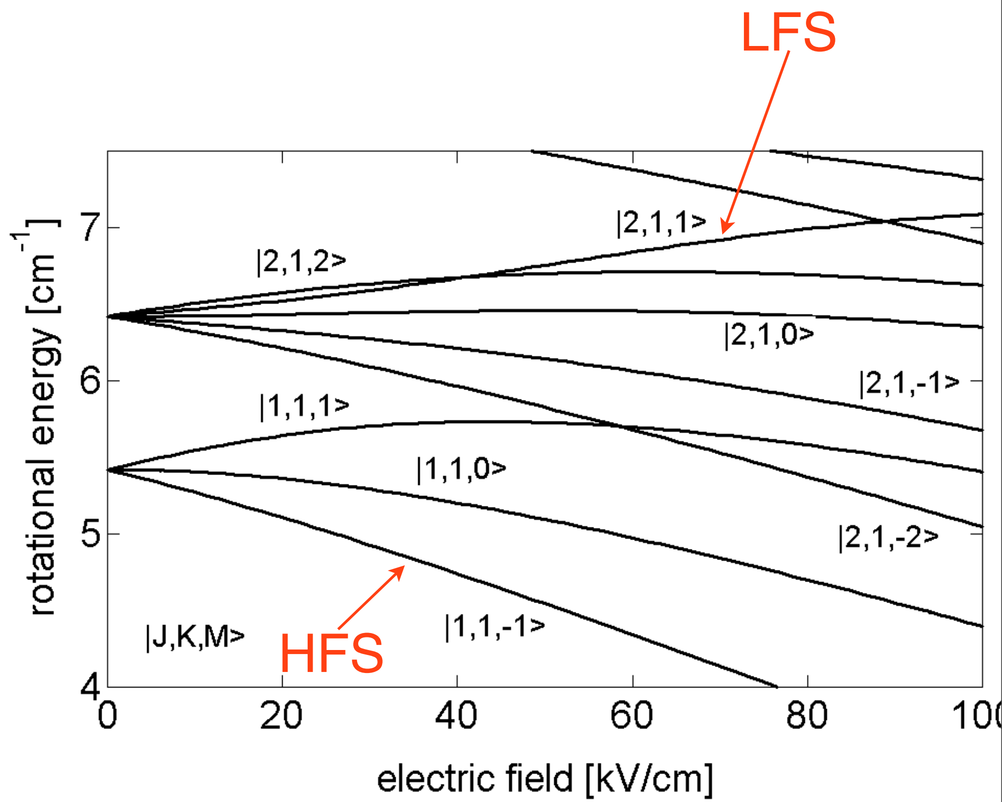

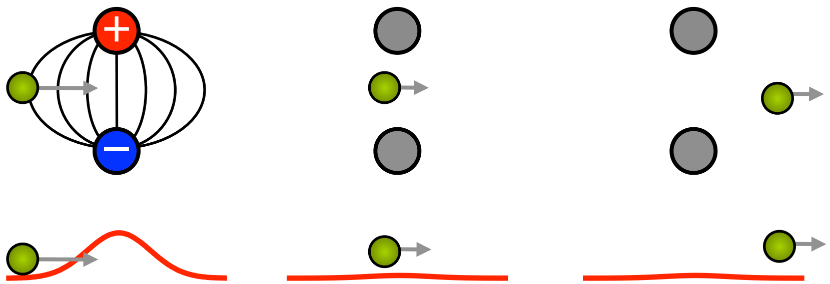

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Zeeman vs Stark Effects

	Stark	**Zeeman**
Hamiltonian	$\hat{H}_S = -\mu \cdot \mathbf{E}$	$\hat{H}_Z = -\mu \cdot \mathbf{H}$
dipole moment	electric dipole 1 D = 3.3564×10^{-30} Cm	magnetic dipole 1 $\mu_B = 9.274 \times 10^{-24}$ JT^{-1}
field	electric 20 kV / 5 mm = 4 MV m^{-1}	magnetic 10 T
shift	$\mu E = 9.72 \times 10^{-5}$ aJ = 4.9 cm^{-1} = 7.0 K	$\mu_B H = 9.28 \times 10^{--5}$ aJ = 4.6 cm^{-1} = 6.8 K

Magnetic Field

Free radicals have a large permanent magnetic moment due to an unpaired electron.

Hamiltonian

$$\hat{H} = \hat{H}_R + \hat{H}_{SR} + \hat{H}_{hf} + \hat{H}_Z$$

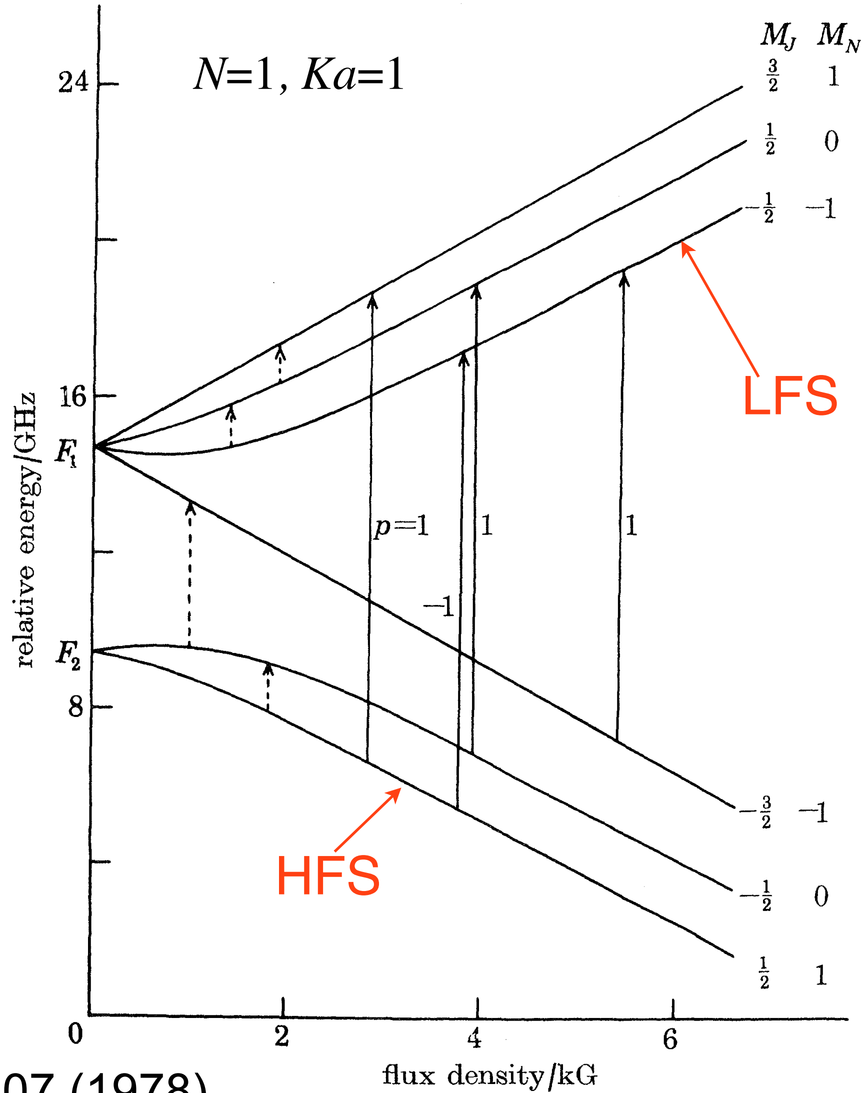
Zeeman term

$$\hat{H}_Z = \mu\,(g_\ell \mathbf{L} + g_S \mathbf{S} + g_I \mathbf{I} + g_N \mathbf{N}) \cdot \mathbf{H}$$

At high field, spin is well decoupled

$$|N M_N K\rangle |S M_S\rangle$$

HCO Zeeman splitting



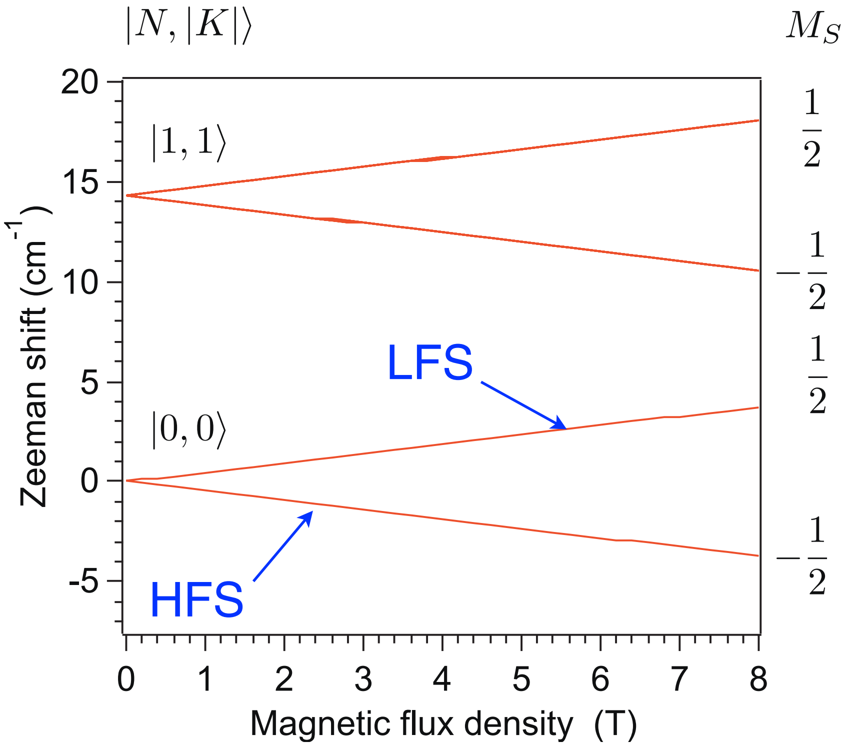
Boland et al.
Proc. R. Soc. Lond. A 360, 507 (1978).

Zeeman Decelerator vs Stark Decelerator

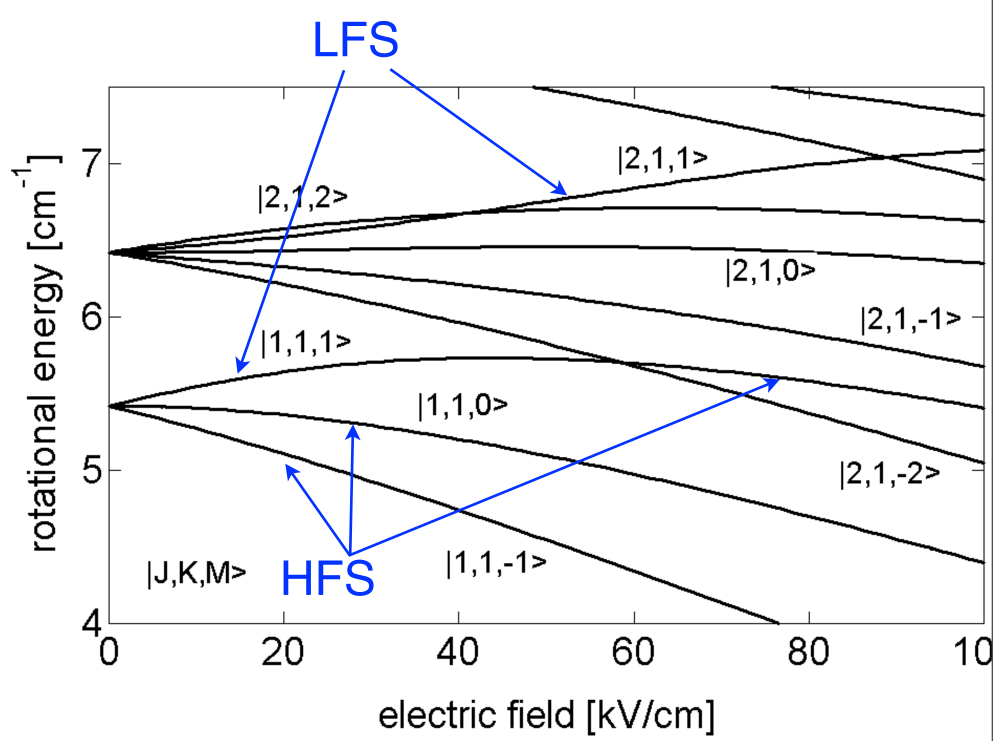
$$|N M_N K\rangle |S M_S\rangle$$

Only LFS are decelerated

Zeeman splitting of CH_3



Stark shift/splitting of CH_3I

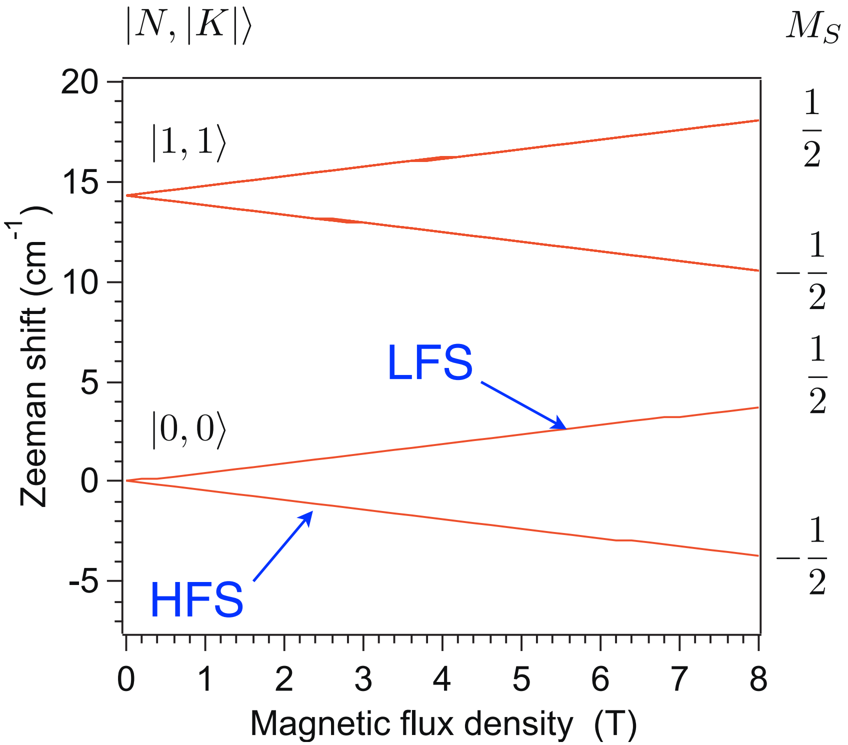


Zeeman Decelerator vs Stark Decelerator

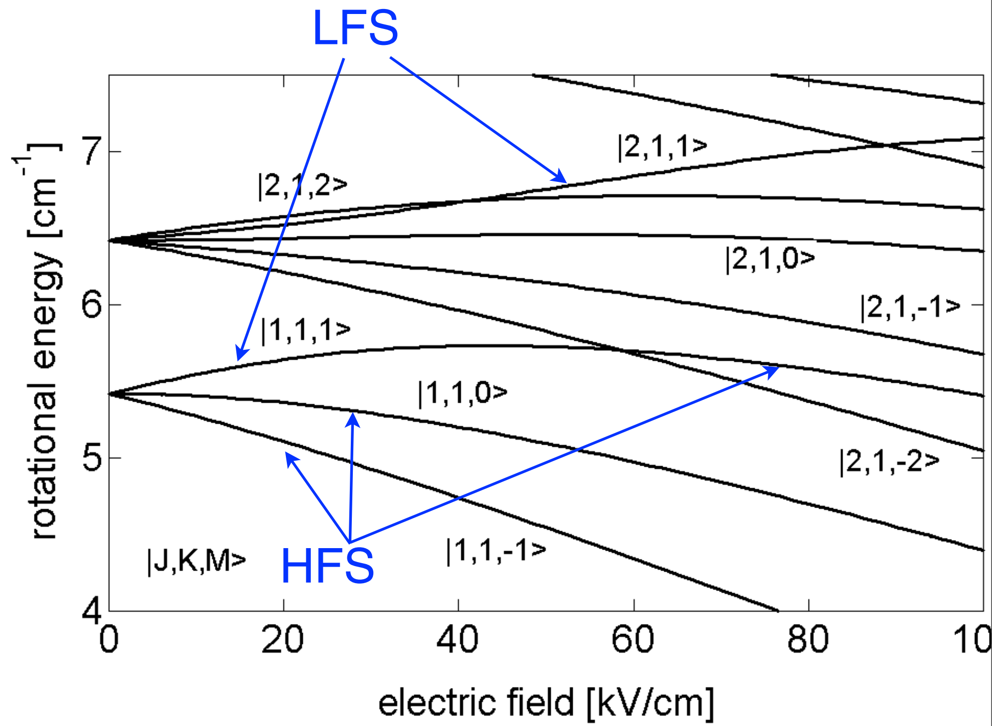
$$|N M_N K\rangle|S M_S\rangle$$

Only LFS are decelerated

Zeeman splitting of CH_3



Stark shift/splitting of CH_3I



Lower rotational states become HFS at high fields

The rotational ground state cannot be decelerated.

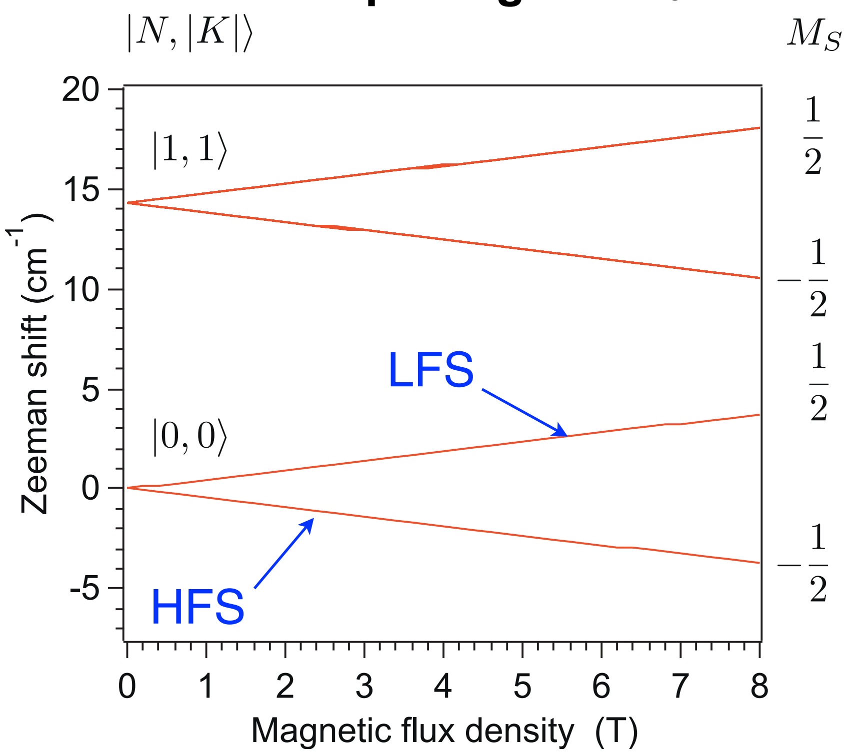
Only small molecules

Zeeman Decelerator vs Stark Decelerator

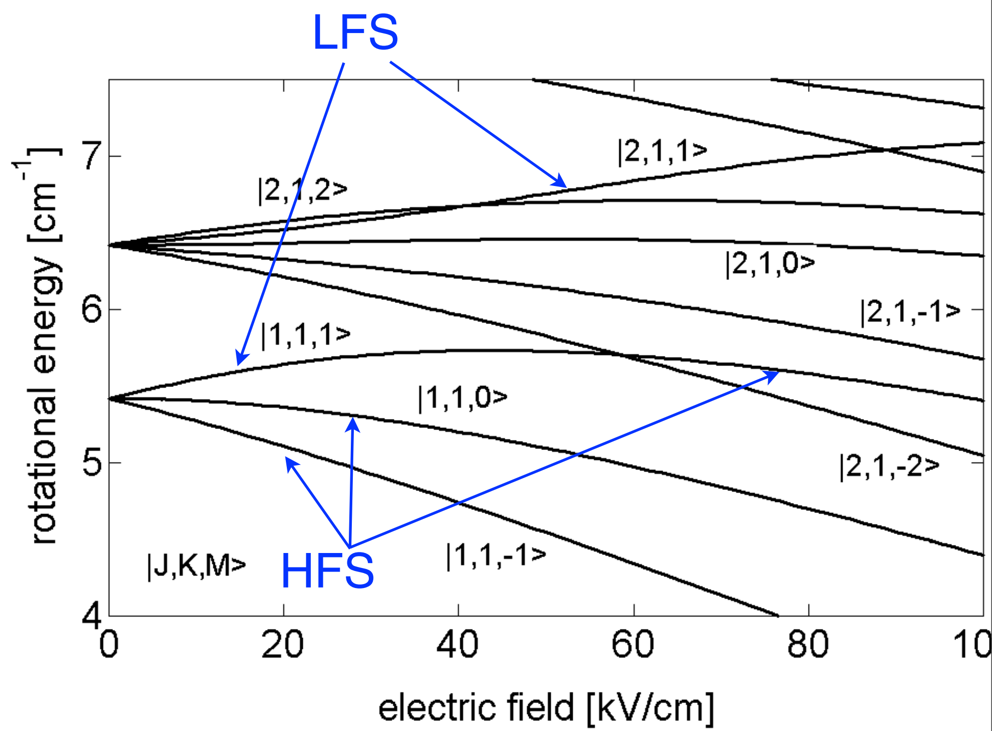
$$|N M_N K\rangle |S M_S\rangle$$

Only LFS are decelerated

Zeeman splitting of CH_3



Stark shift/splitting of CH_3I



Any ground rotational state has a LFS component.

The ground rotational state can be decelerated.

More universal

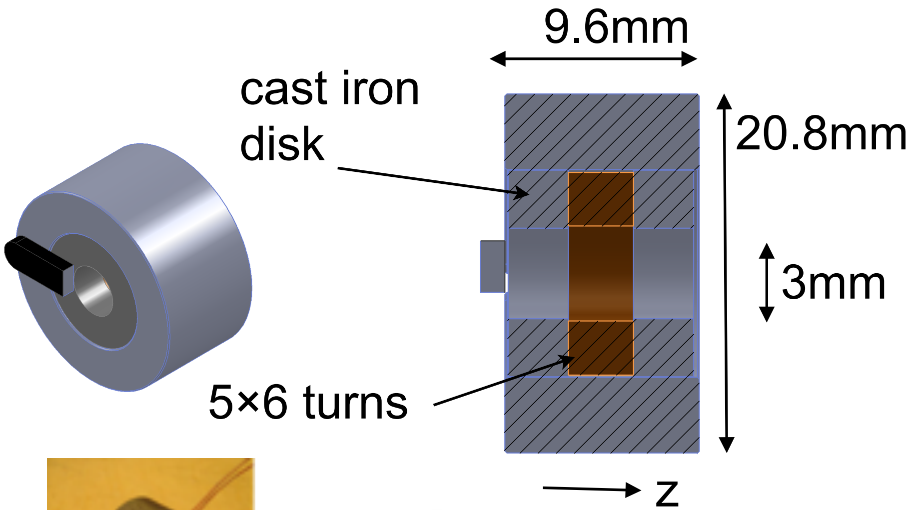
Lower rotational states become HFS at high fields

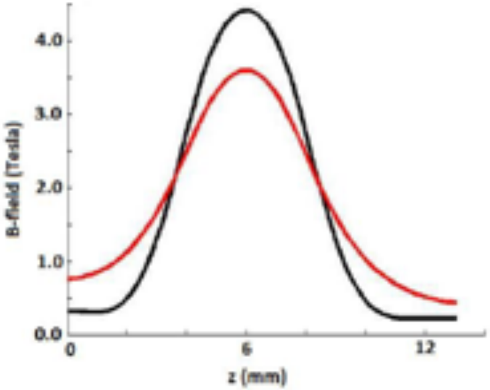
The rotational ground state cannot be decelerated.

Only small molecules

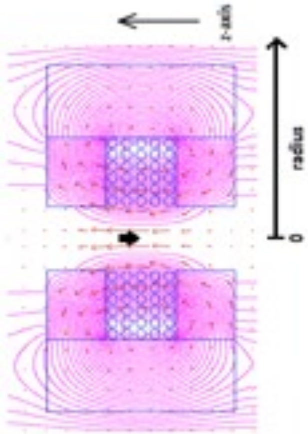
High Field Solenoid Coil @ CRUCS

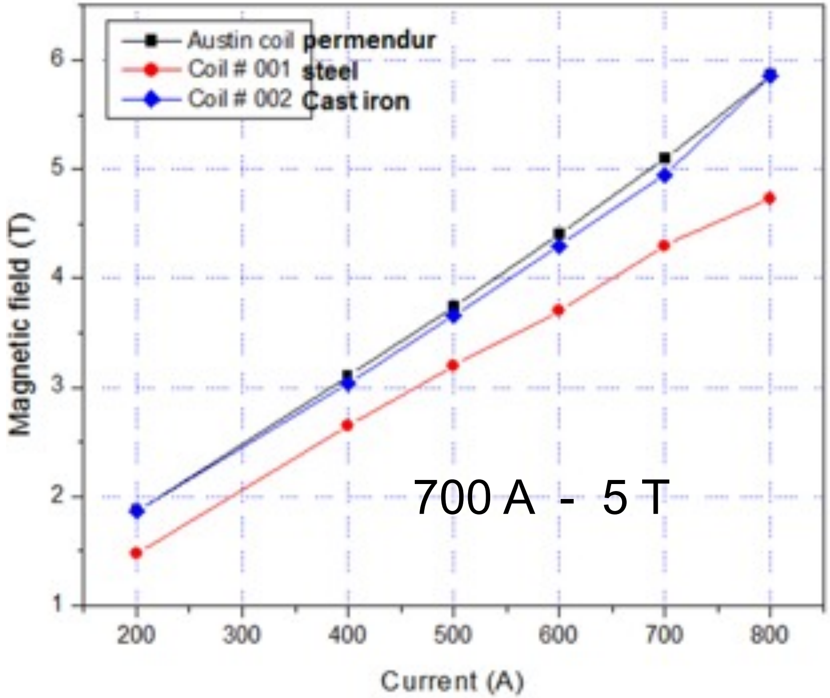
30 turns Solenoid coil





M. Raizen (Texas, Austin)





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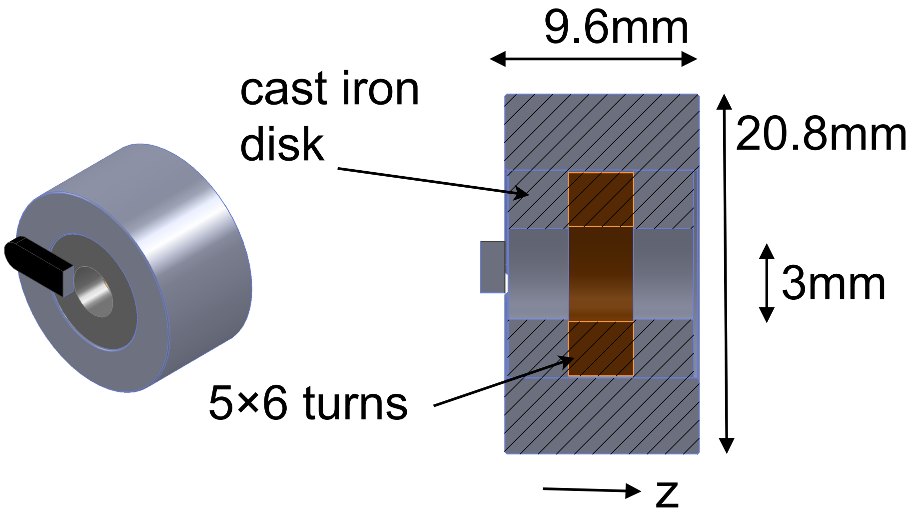

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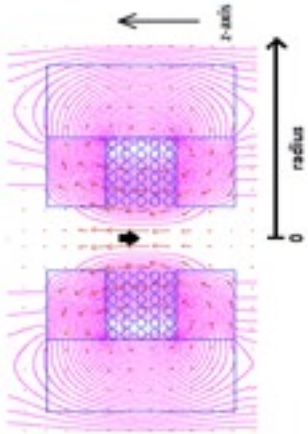
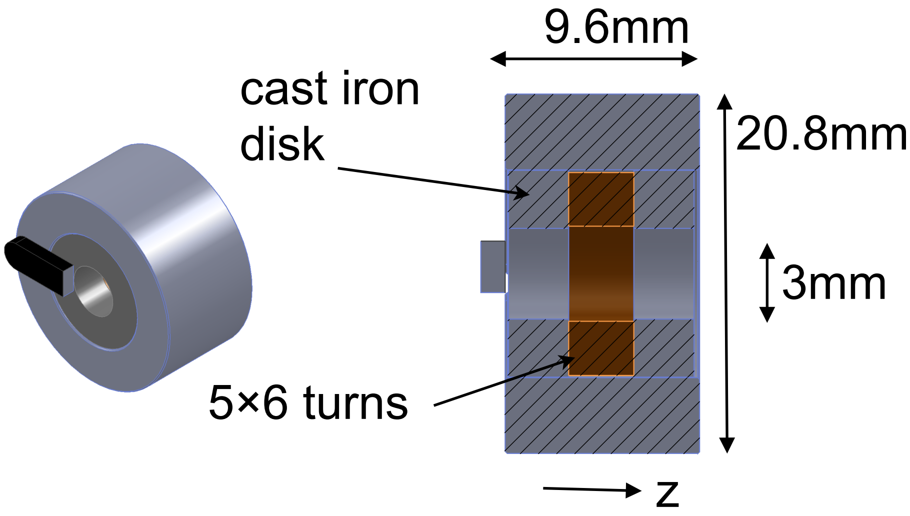


v=400 m/s → 4μs/1.5 mm

High Field Solenoid Coil @ CRUCS

30 turns Solenoid coil



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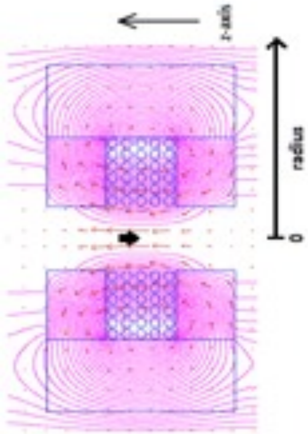

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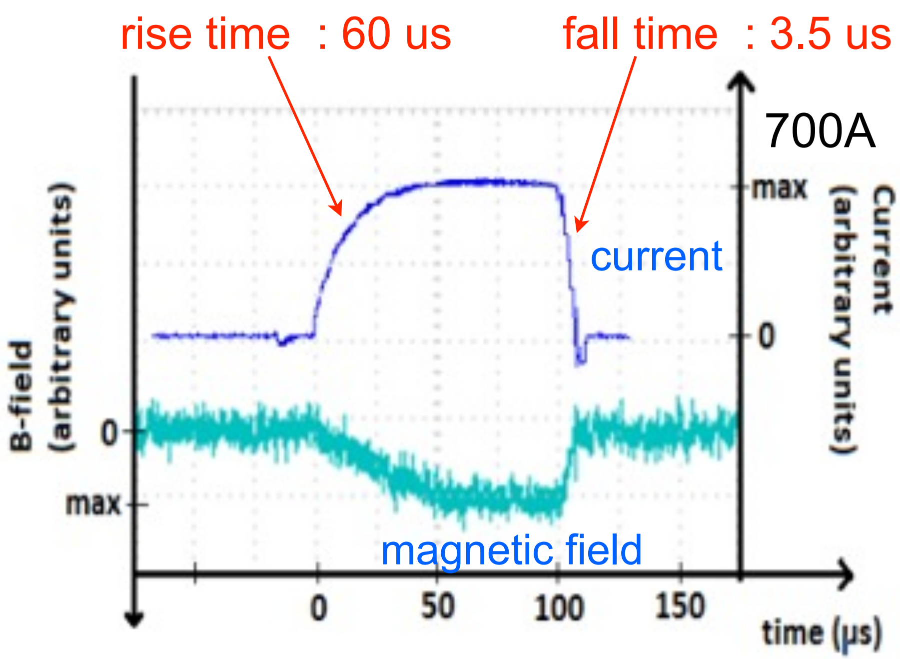
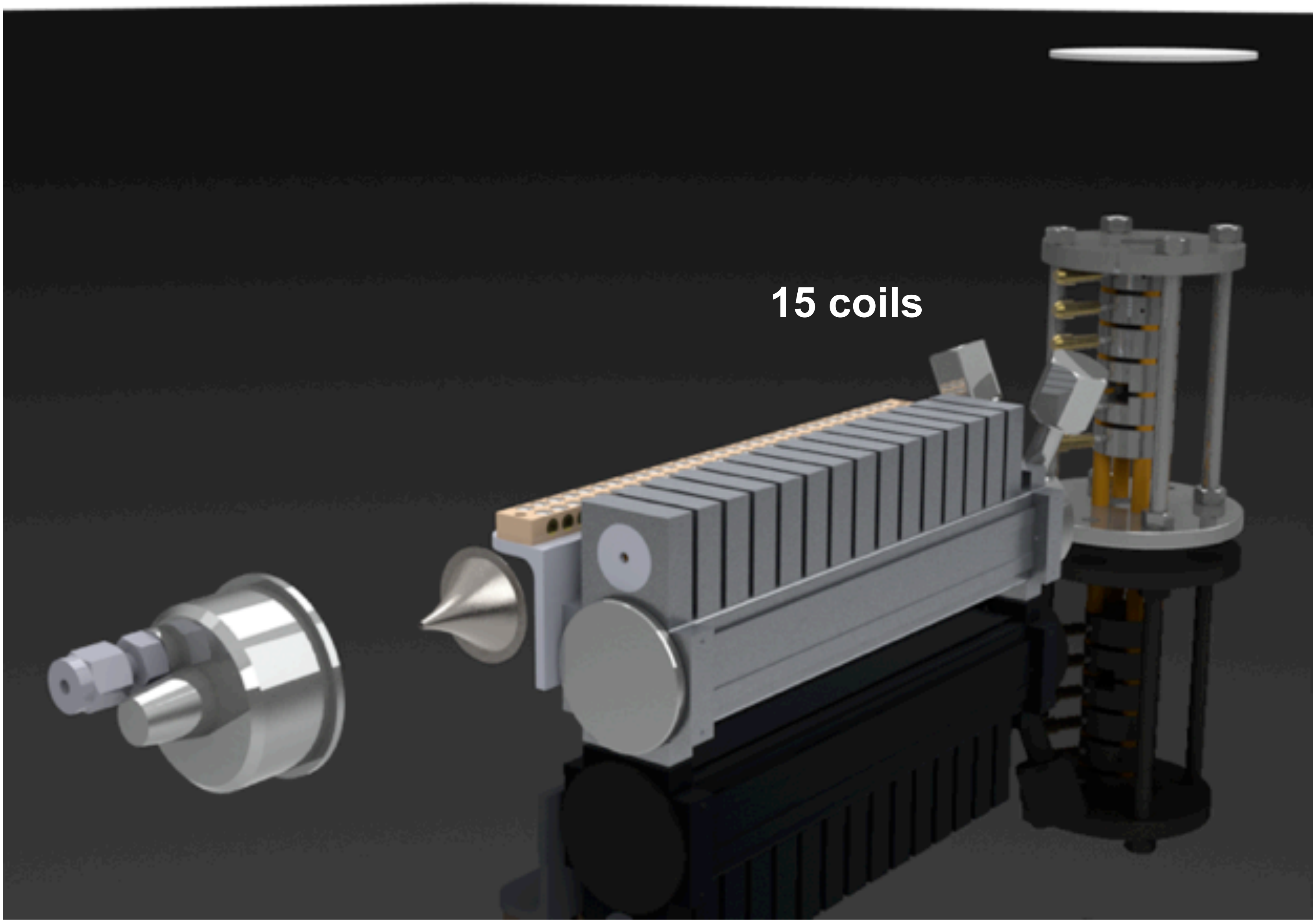
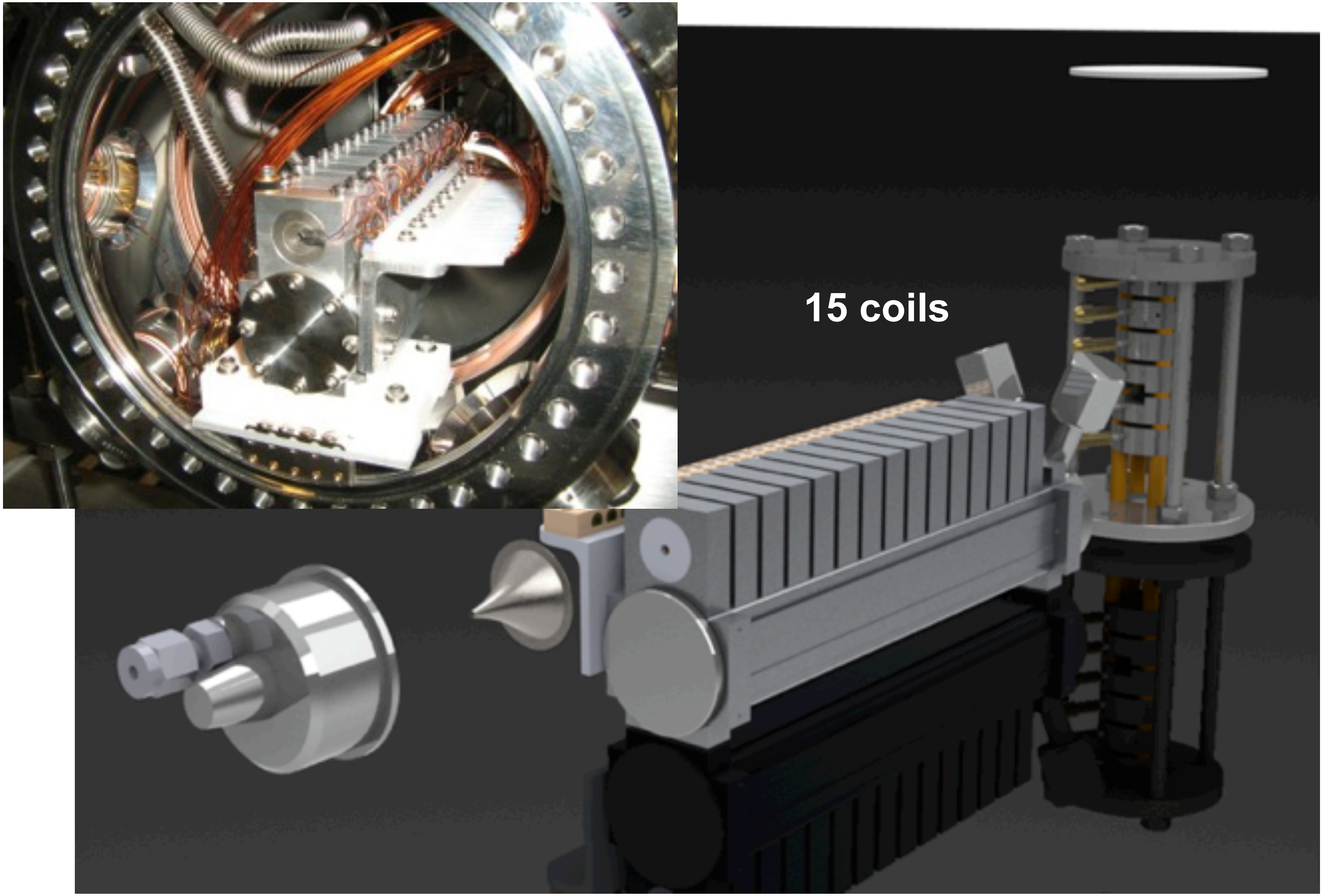


1st Generation Zeeman Decelerator



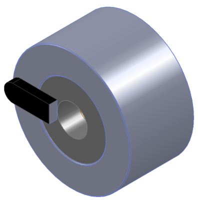
Phys. Chem. Chem. Phys **15**, 1772 (2013)

1st Generation Zeeman Decelerator



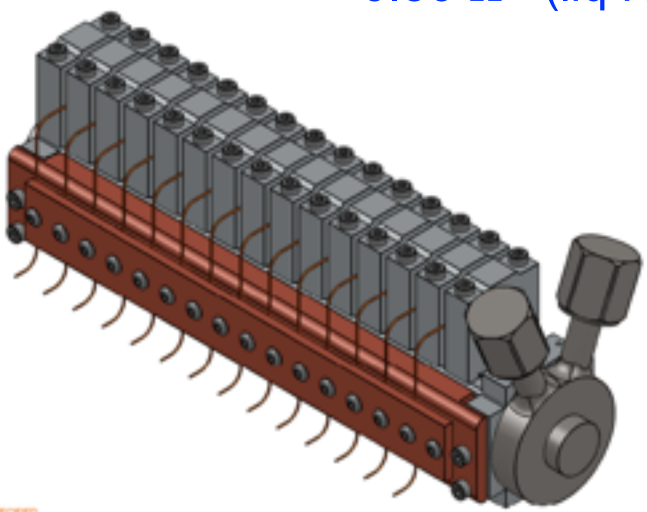
Phys. Chem. Chem. Phys **15**, 1772 (2013)

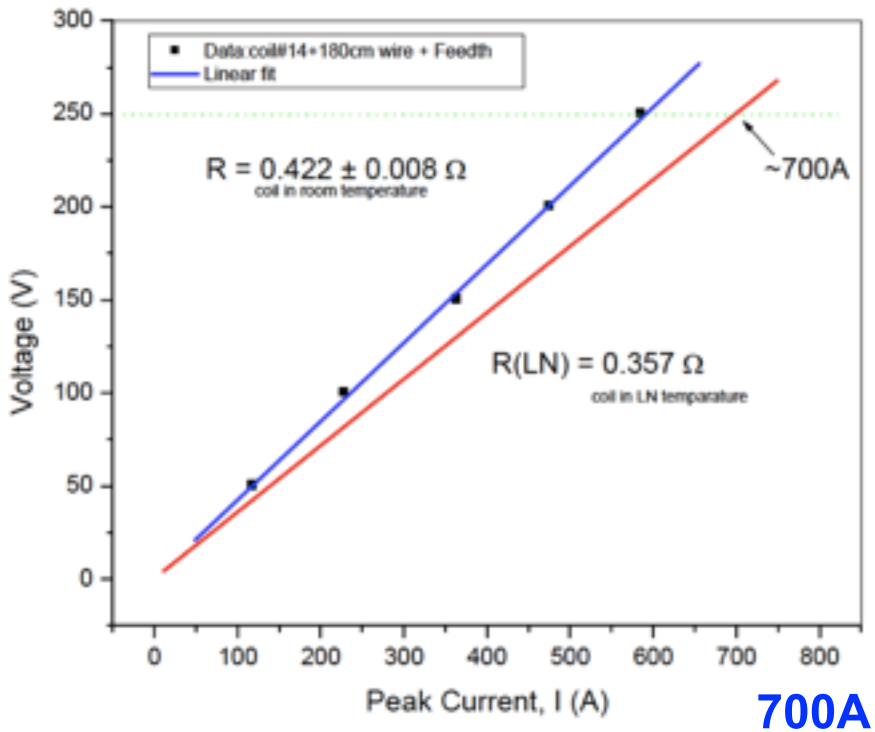
1st Generation Zeeman Decelerator



Liq. N_2 cooling

total resistance
0.42 Ω (RT)
0.36 Ω (liq N_2)

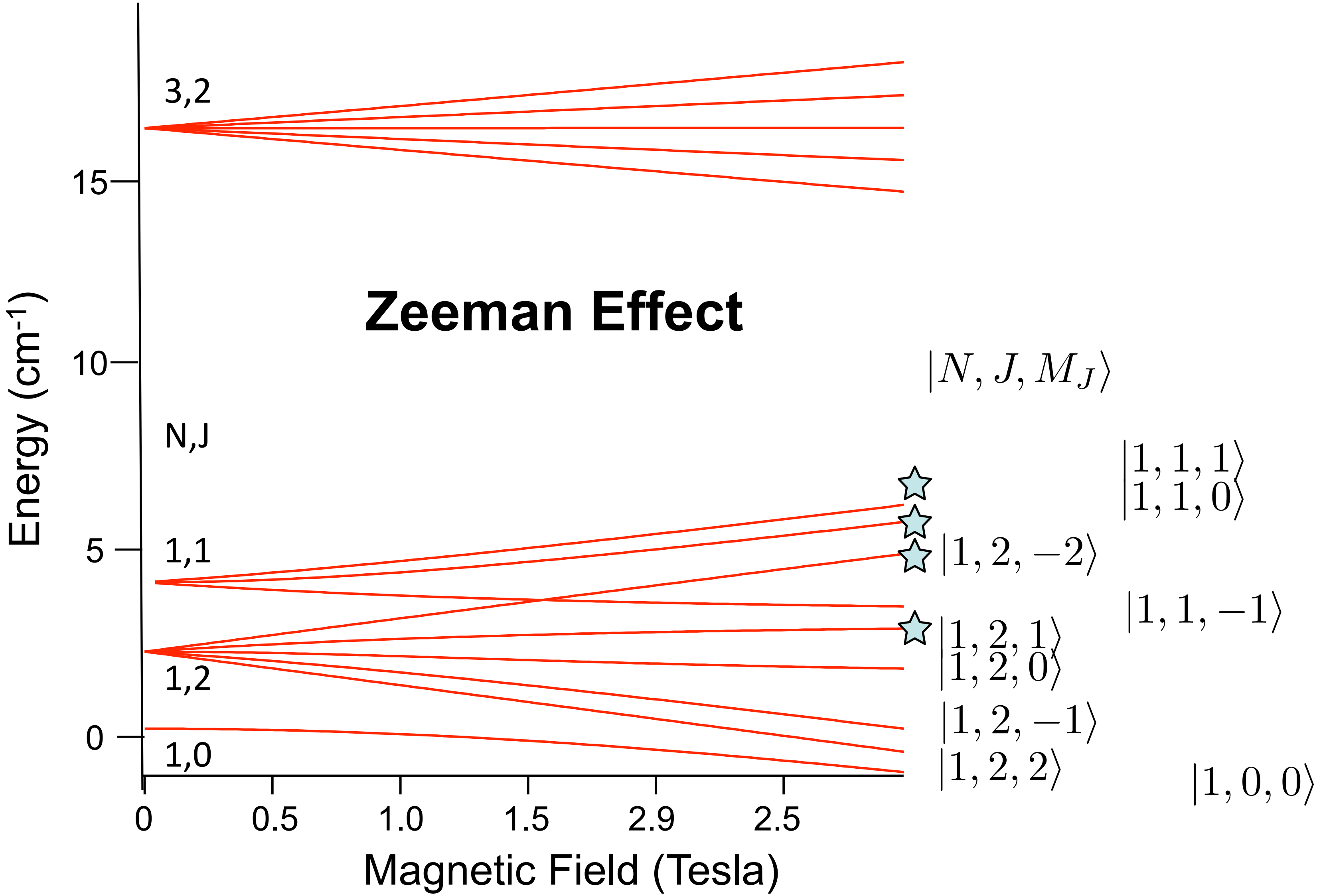




700A : 4.9 T

Test Sample : O_2

$^{16}O_2 \ \ (\tilde{X}^3\Sigma_g^-)$ $\qquad \hat{H} = \hat{H}_R + \hat{H}_{SR} + \hat{H}_{SS} + \hat{H}_Z$



Simulation

$^{16}O_2 \quad |N, J, M_J\rangle = |1, 2, 1\rangle$

initial velocity : 250 m/s
400 A switching

transverse distribution

velocity (m/s)

340
320
300
280
260
240
220
200
180
160

molecular
distribution

longitudinal
distribution

coil switching

1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 **15 coils**

Simulation

$^{16}O_2 \quad |N, J, M_J\rangle = |1, 2, 1\rangle$

initial velocity : 250 m/s
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transverse distribution

molecular distribution

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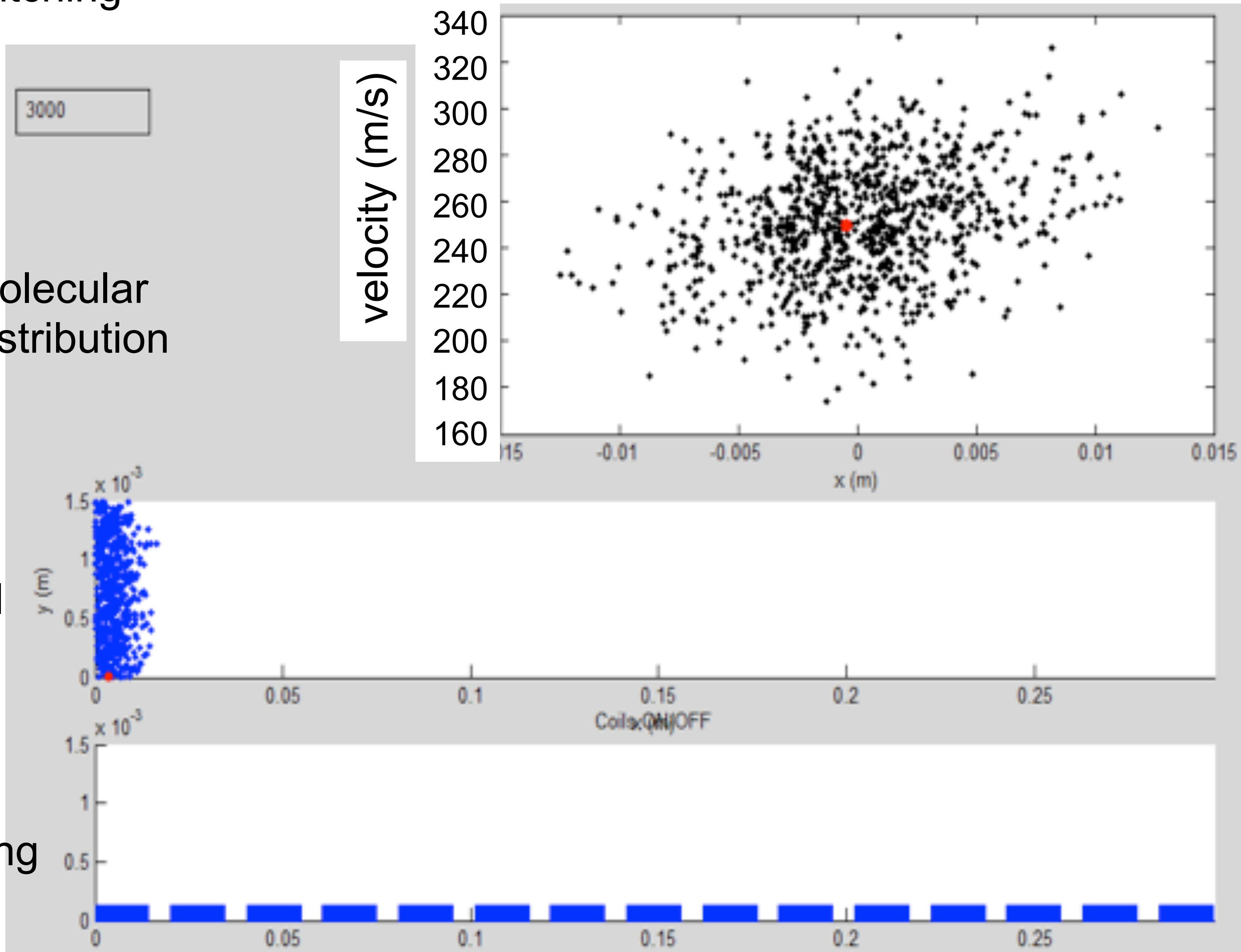
coil switching



1 2 3 4 5 6 7 8 9 10 11 12 13 14 15

15 coils

Simulation

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180
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molecular
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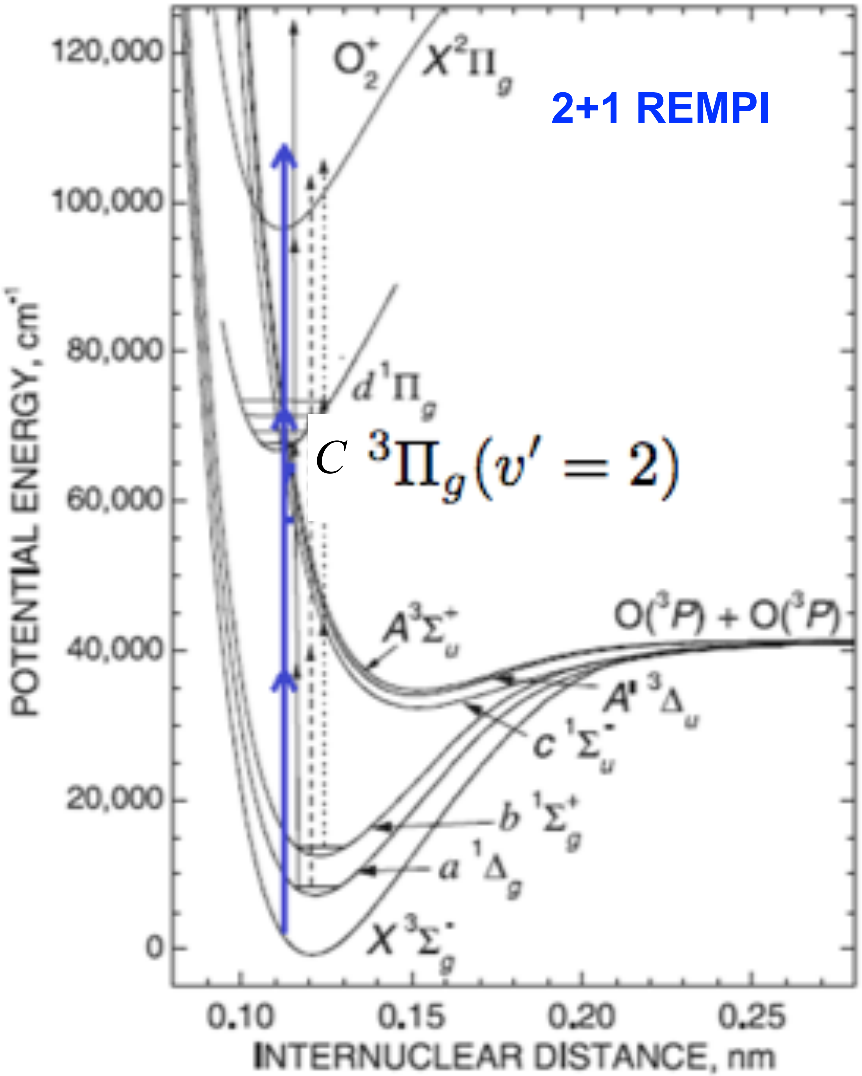
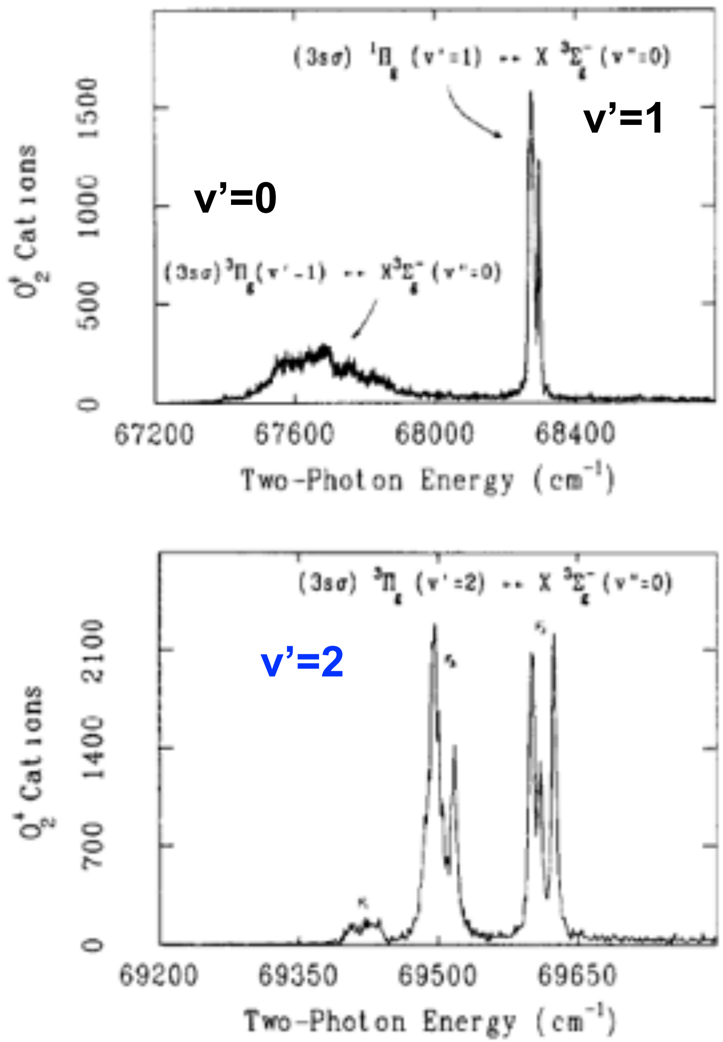
longitudinal
distribution

coil switching

1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 **15 coils**

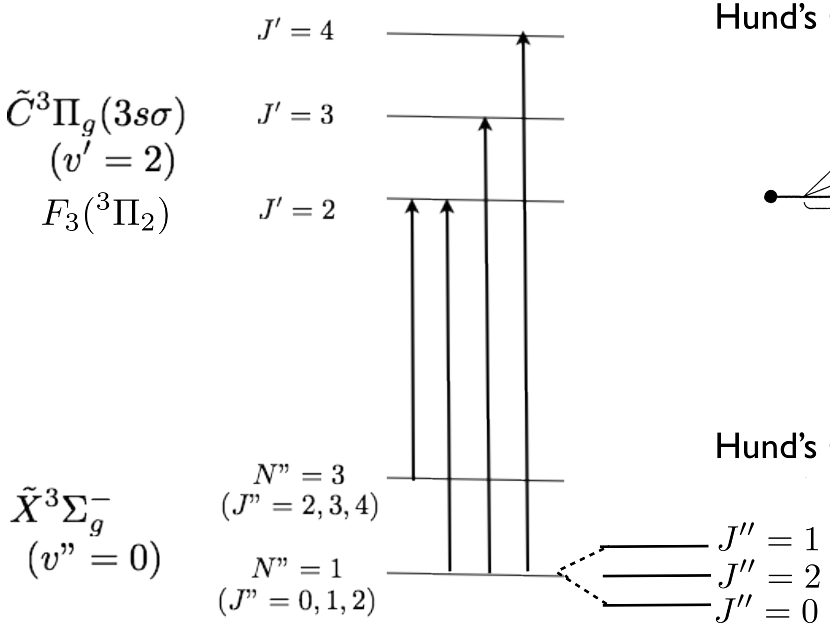
O_2 Detection : 2+1 REMPI



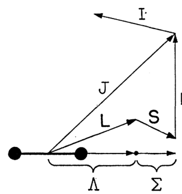


McCann, Chen, Datskou, Evans, Chem Phys. ***174****, 417 (1993)*

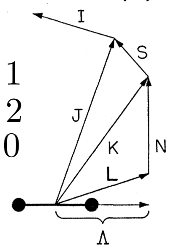
O_2 REMPI Detection



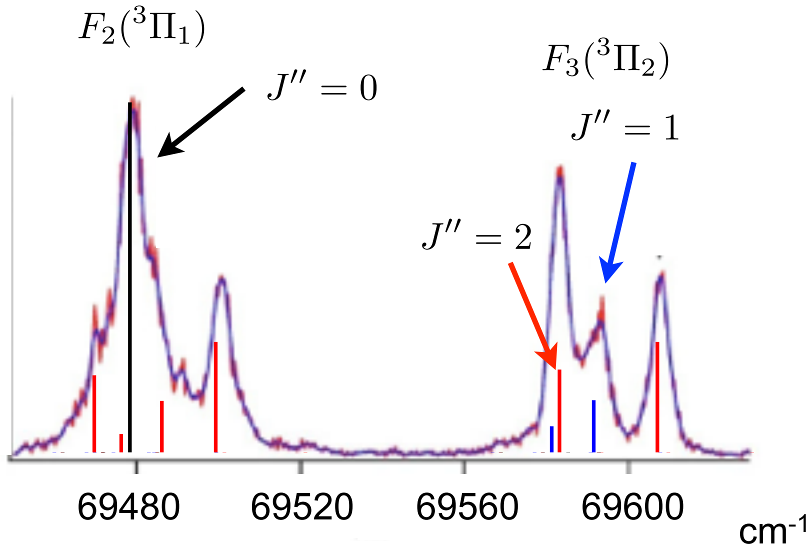
Hund's Case (a)



Hund's Case (b)



J" levels are not completely resolved

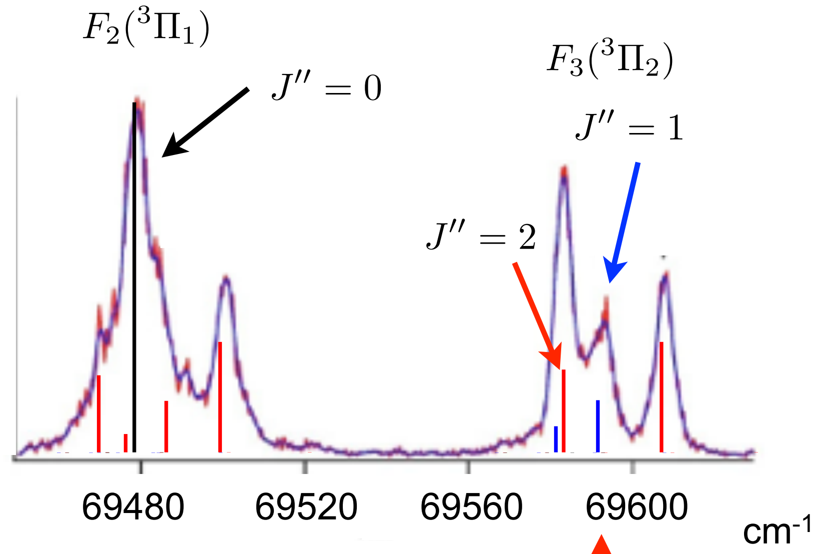


rotational temp   3 - 5 K

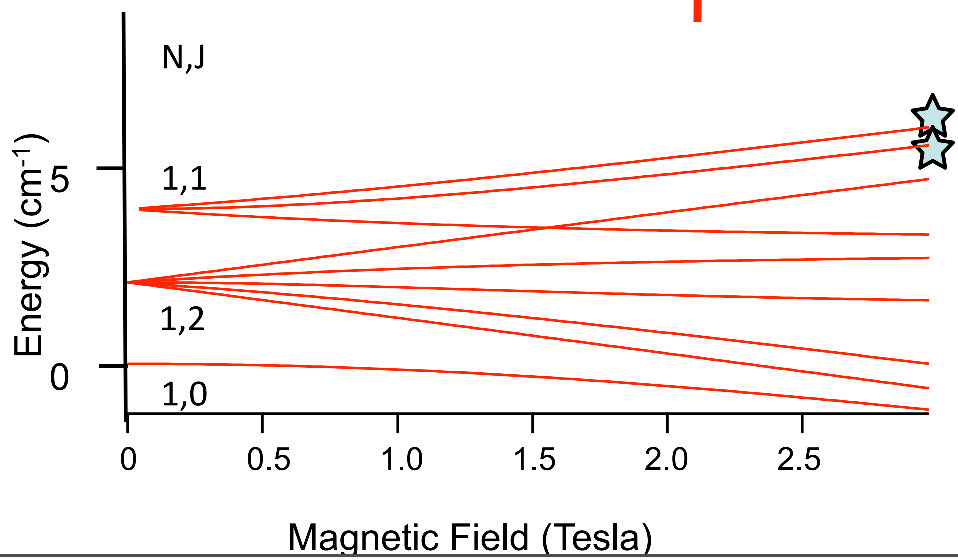
assignments

Katsumata et al.
J. Elec. Spec. Rel. Phenom. **41**, 325 (1986).

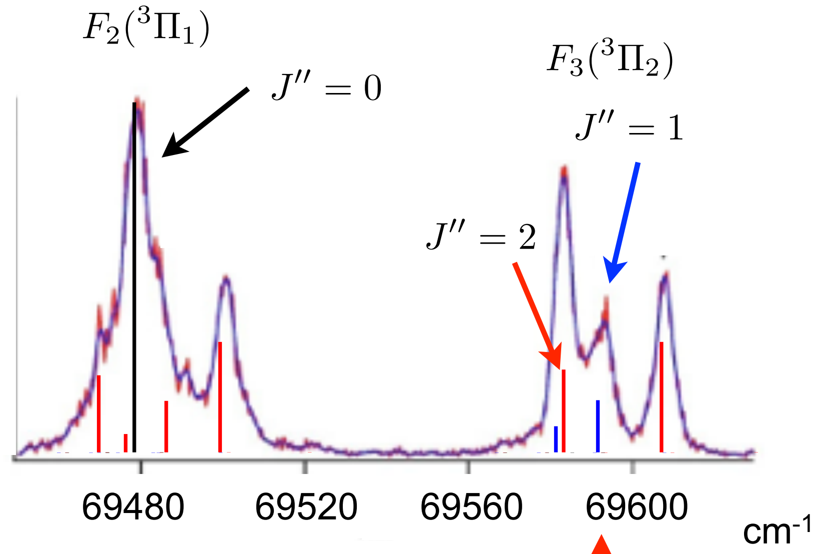
Zeeman Deceleration of O_2



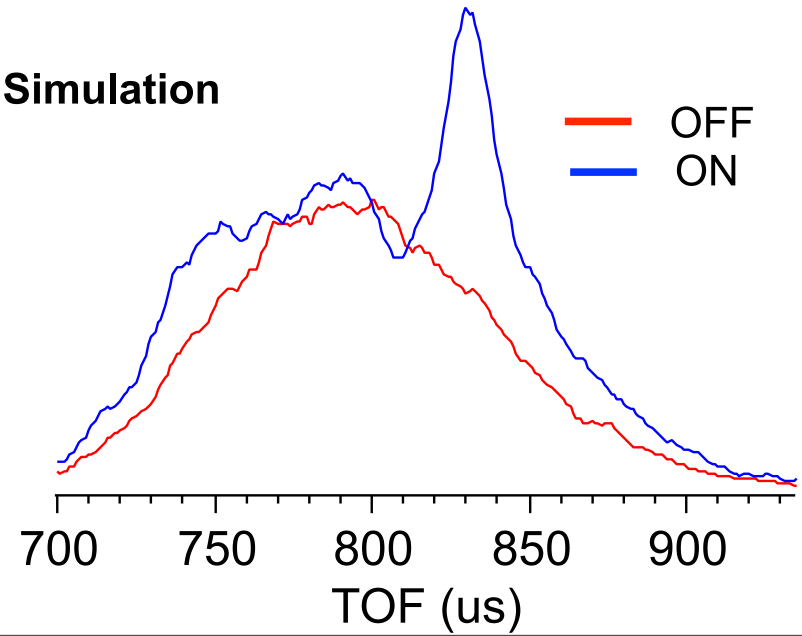
Zeeman shift



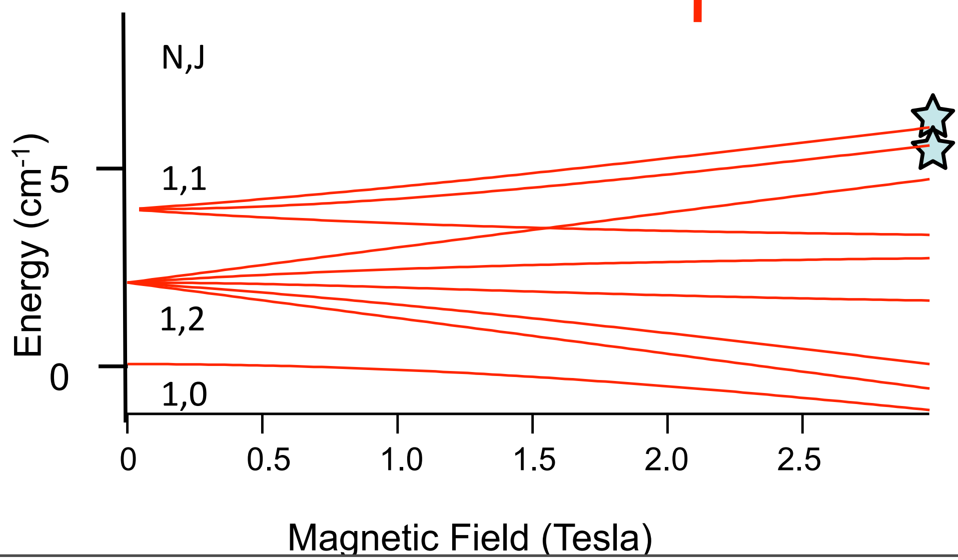
Zeeman Deceleration of O_2



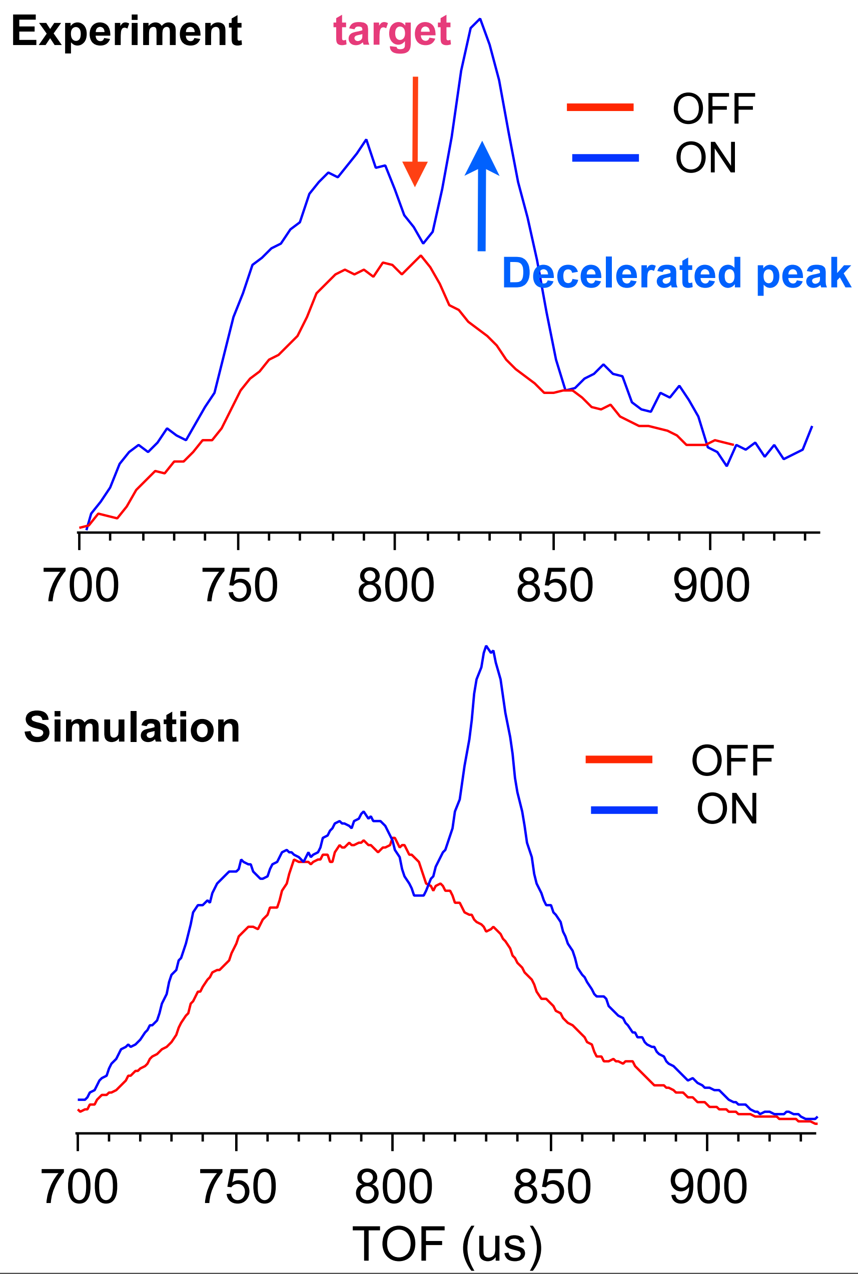
Simulation

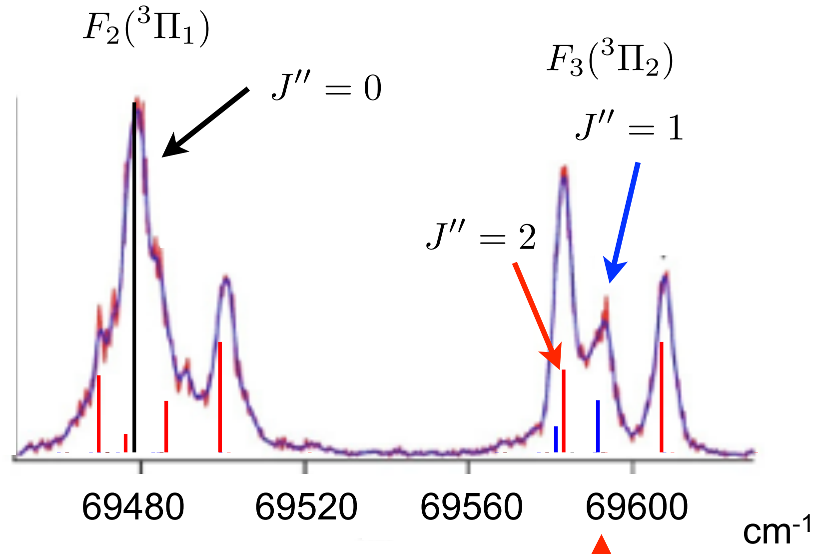


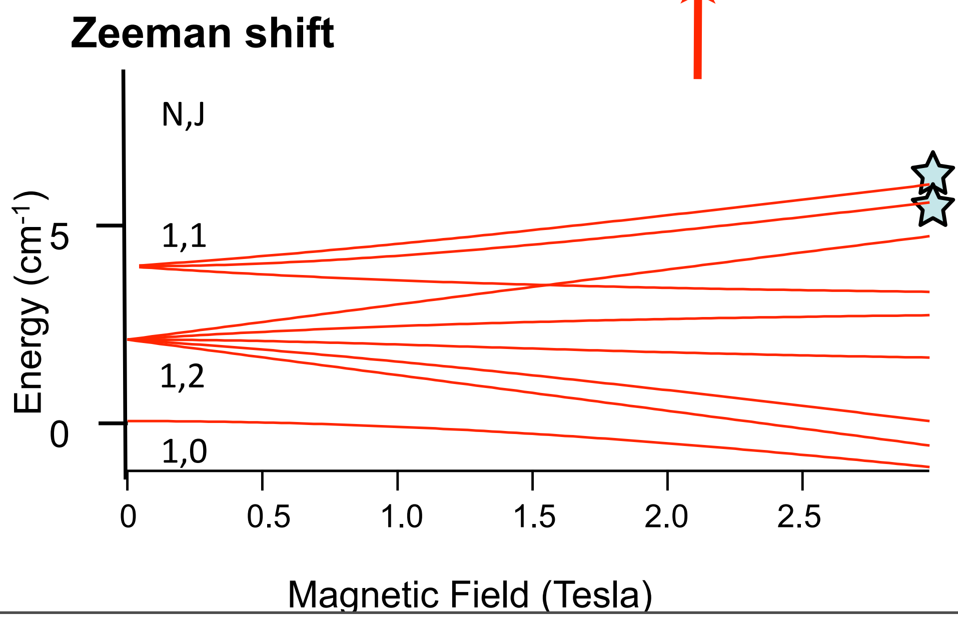
Zeeman shift



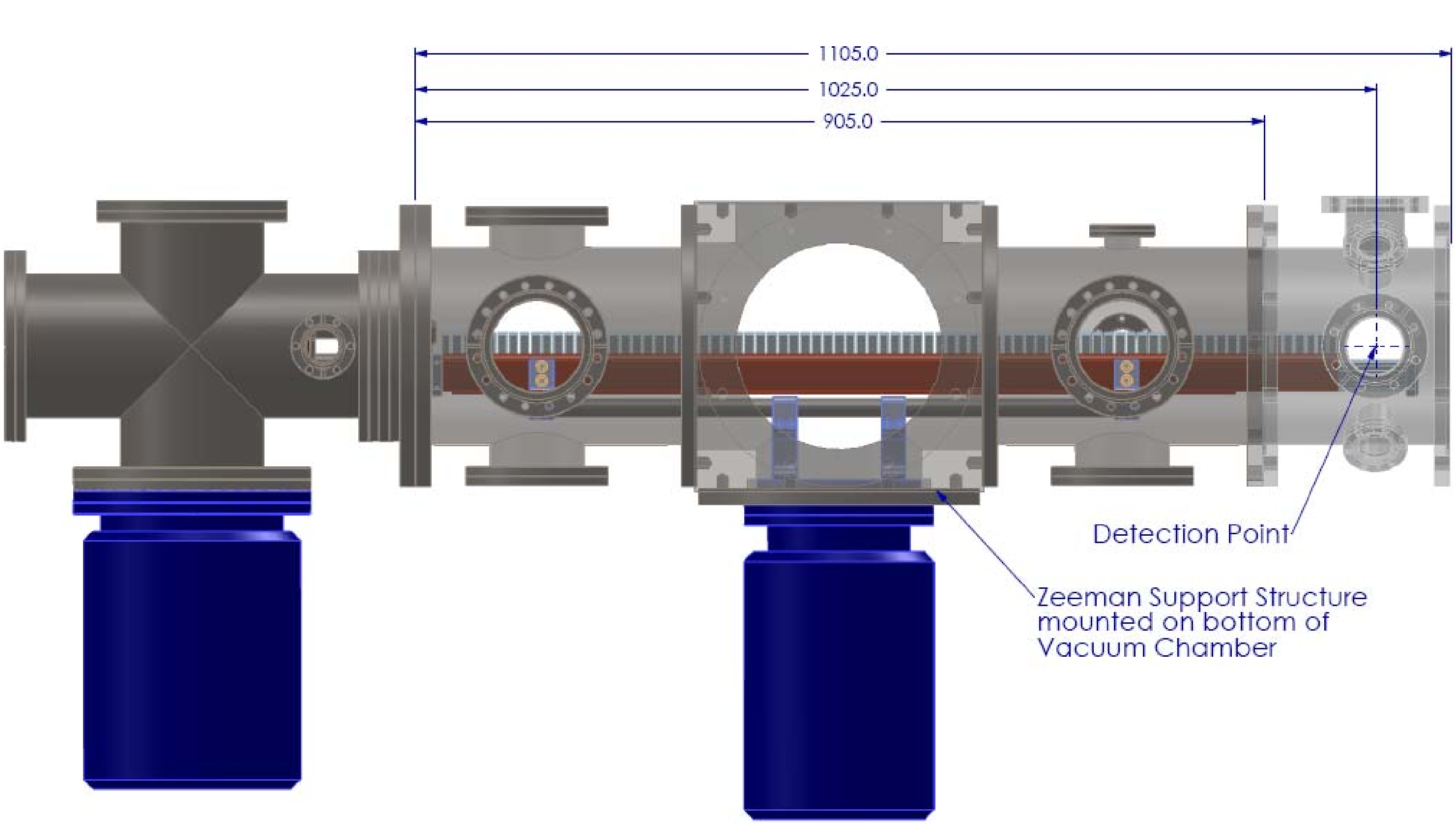
Zeeman Deceleration of O_2



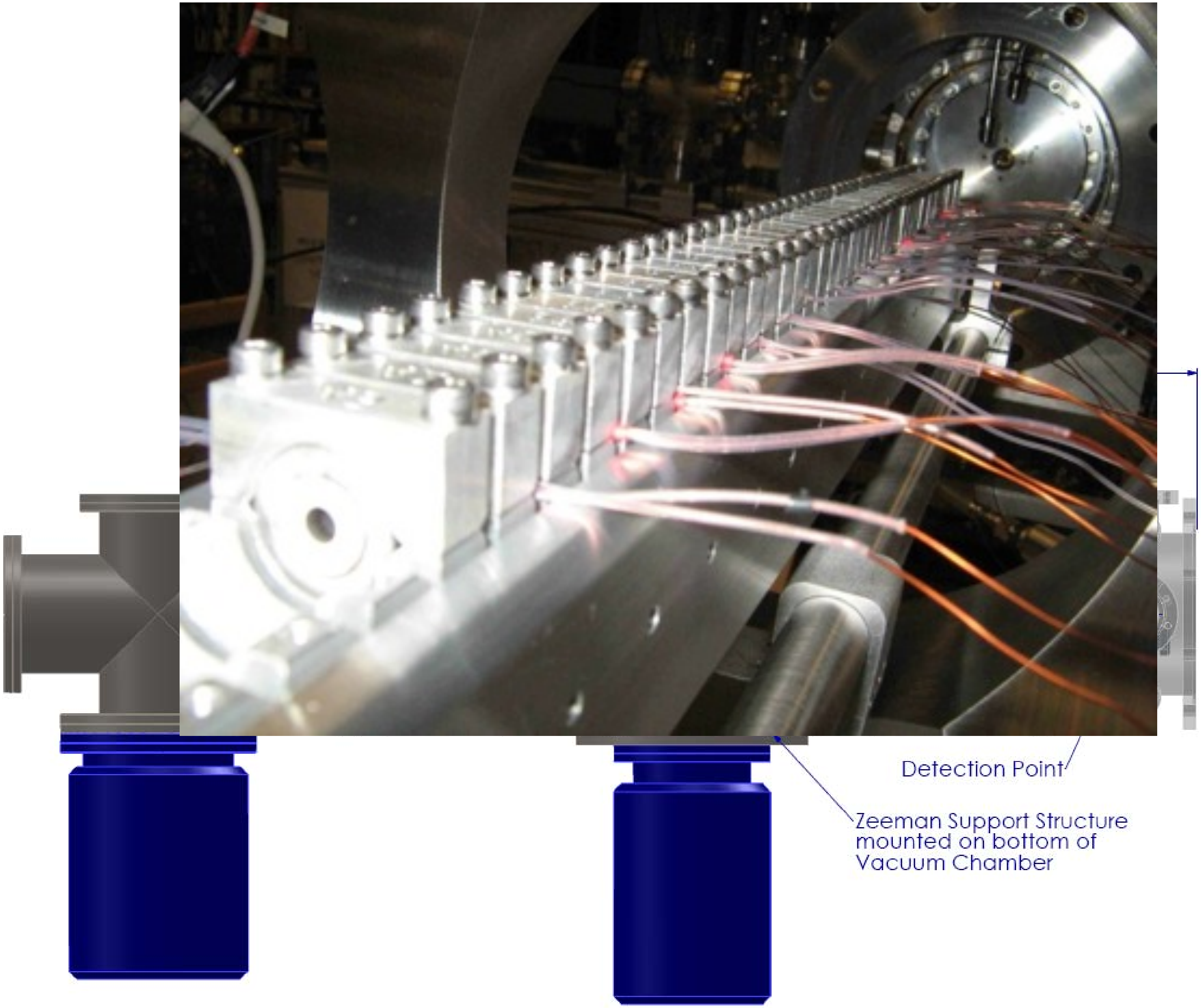




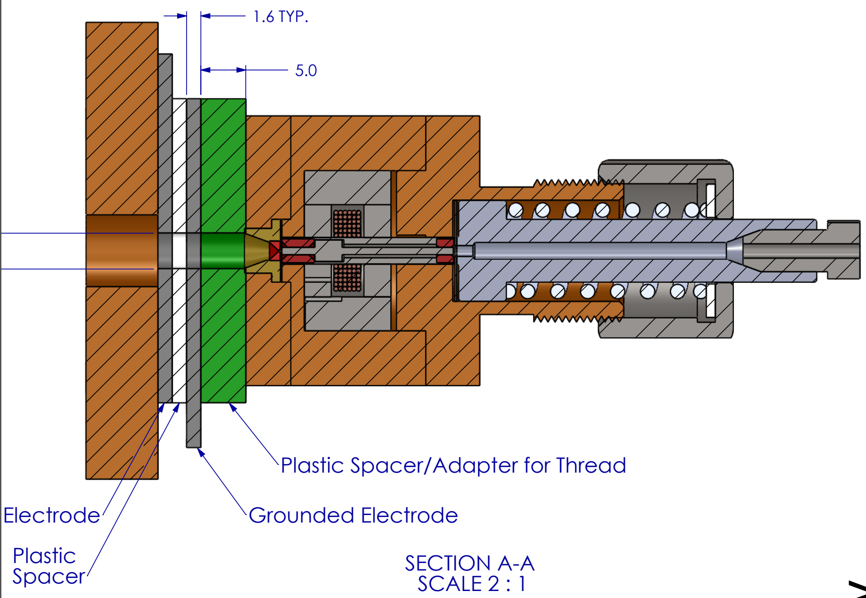
2nd Generation : 80 Coils



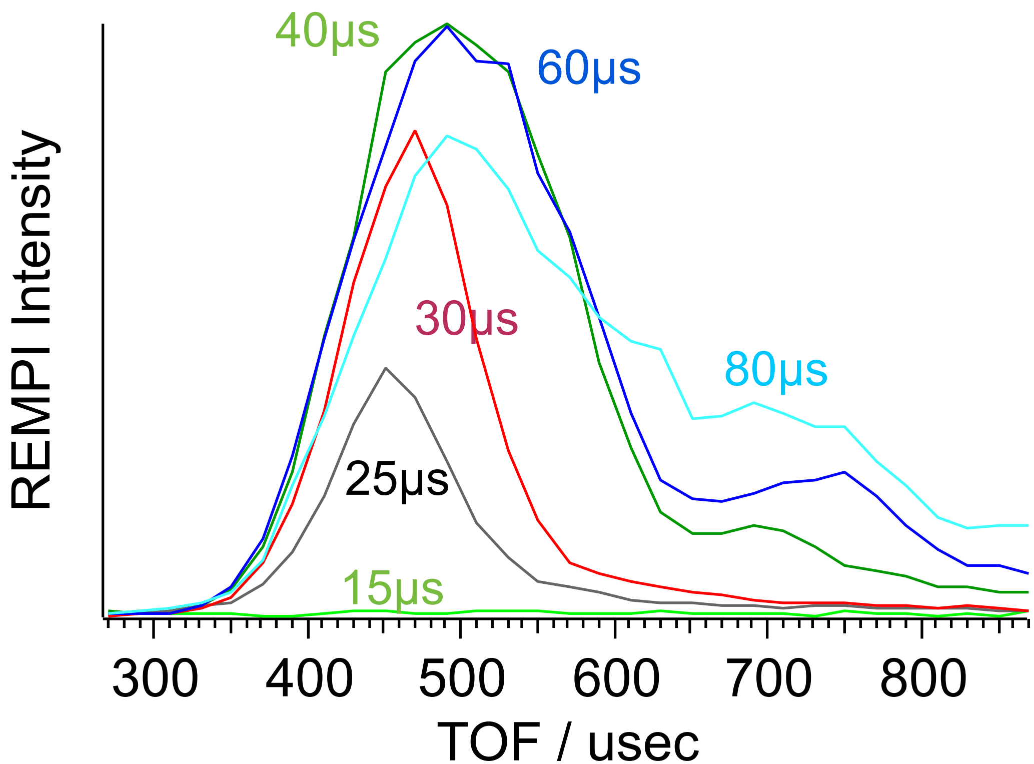
2nd Generation : 80 Coils



“CRUCS” Pulsed Valve

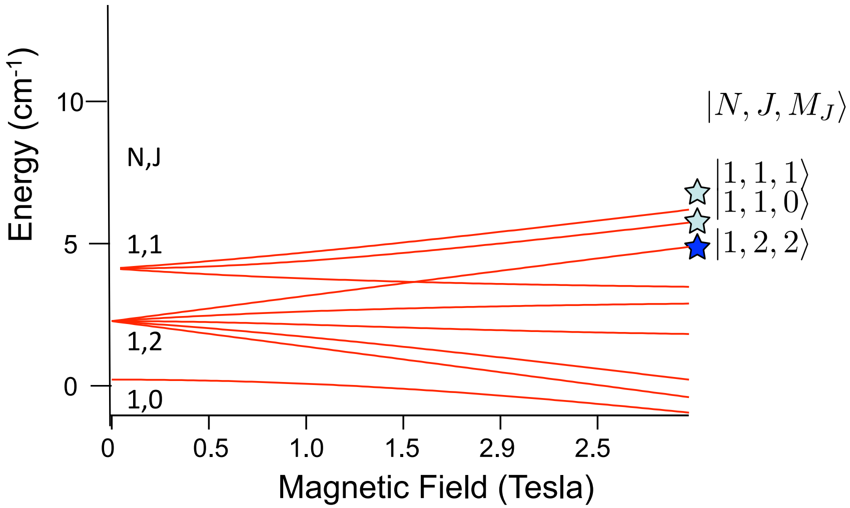


Opening time ~ 25 us
Low & High stagnation pressure (1 - 10 bar)
able to operate at liq. He temp

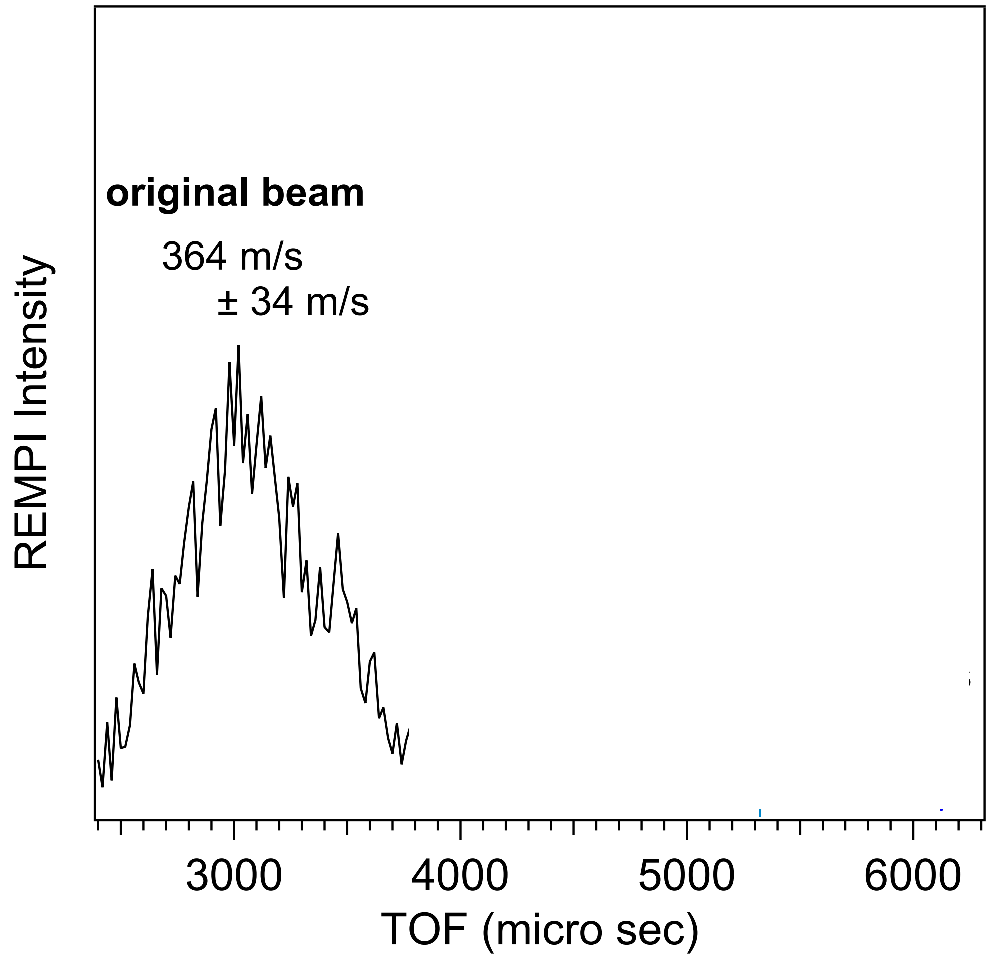


Deceleration of O_2

80-coil system

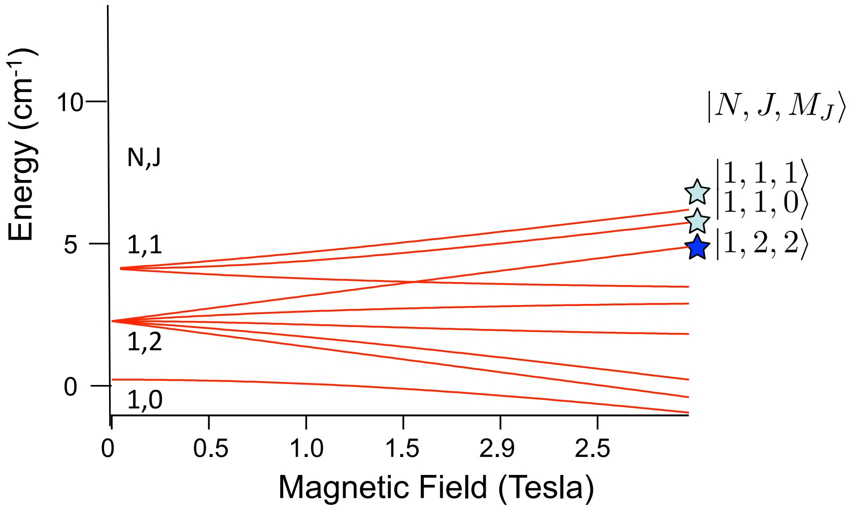


O_2 seeded in Kr @ 135 K (65 us open)



Deceleration of O_2

80-coil system



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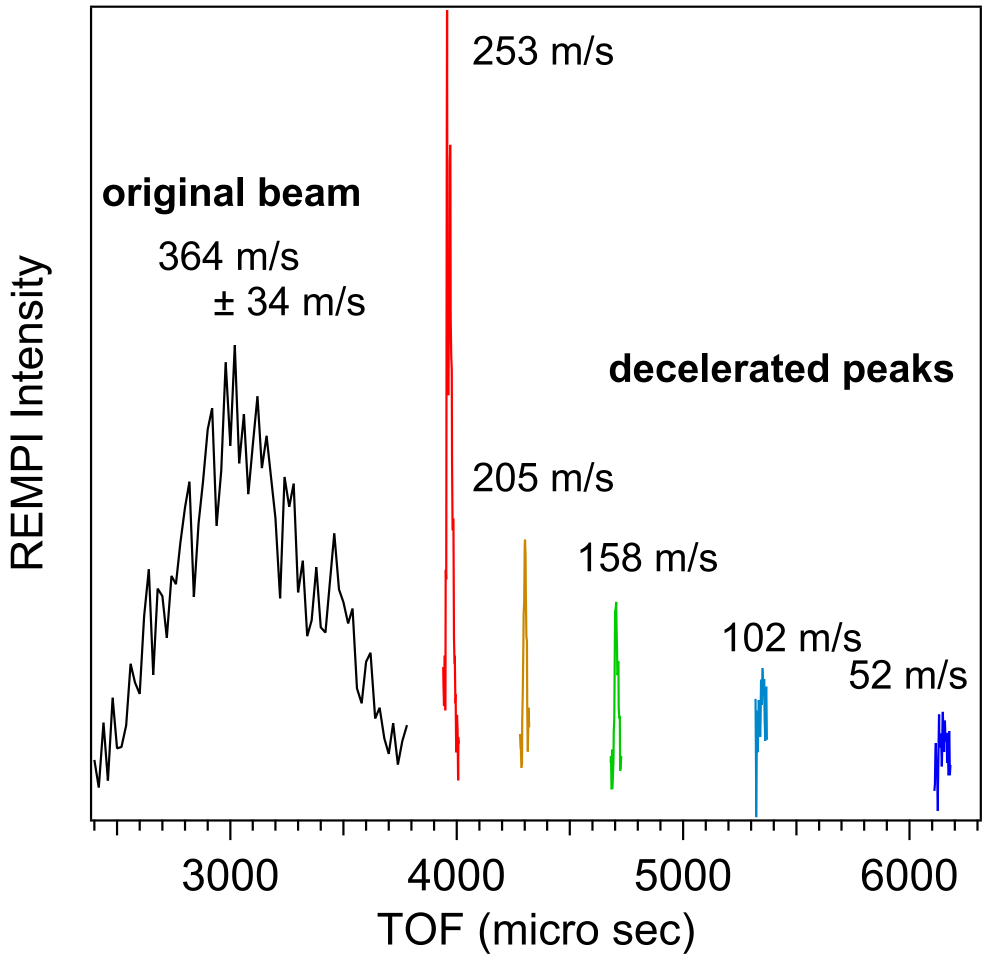

Deceleration of O_2

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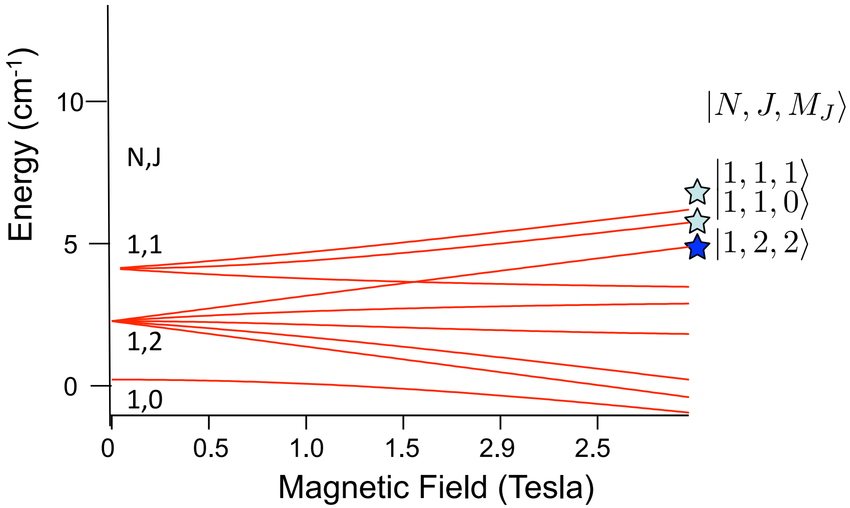

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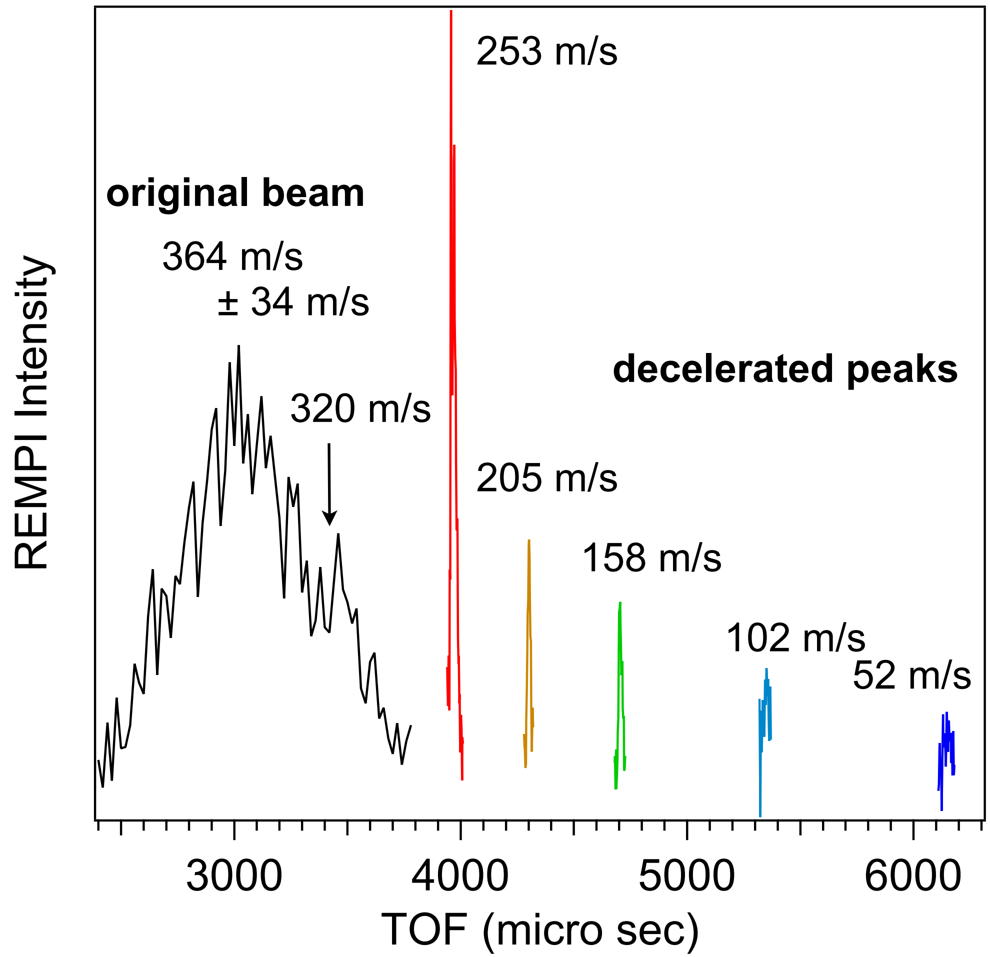
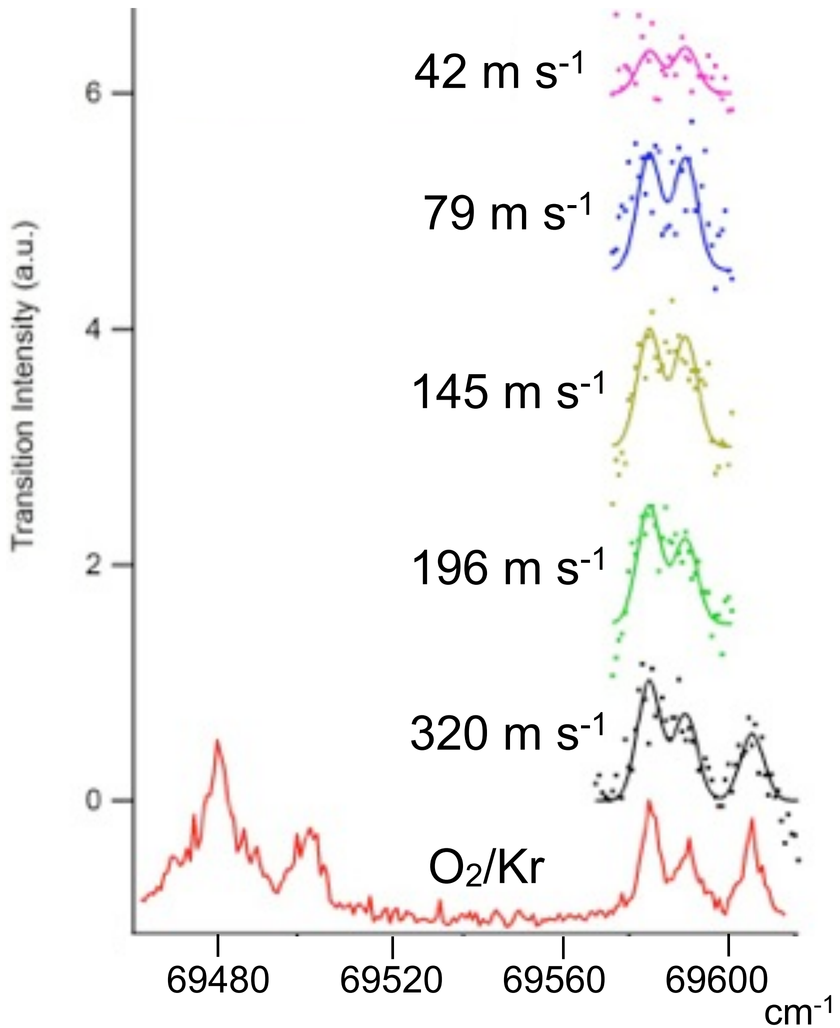


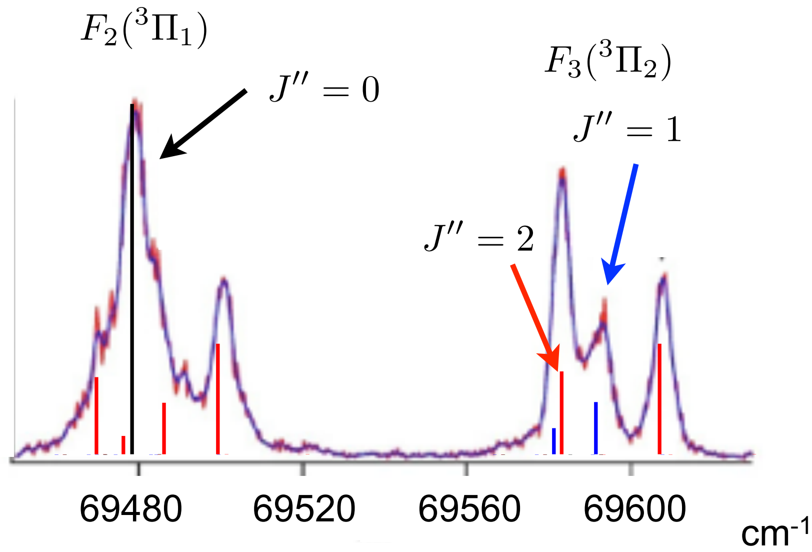
320 m/s ~ 200 K

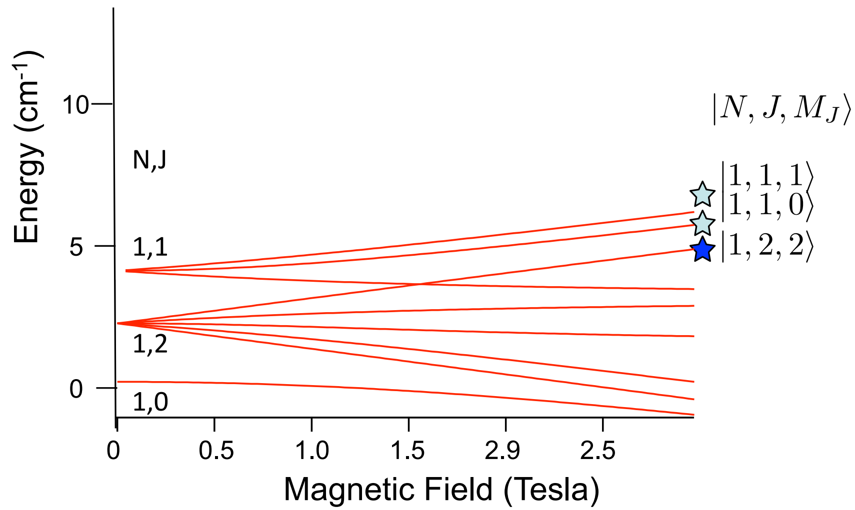
52 m/s ~ 4 K

REMPI Spectra of Decelerated O_2

|J=1, M_J=0,1> (and |J=2,M_J=2>) are decelerated.

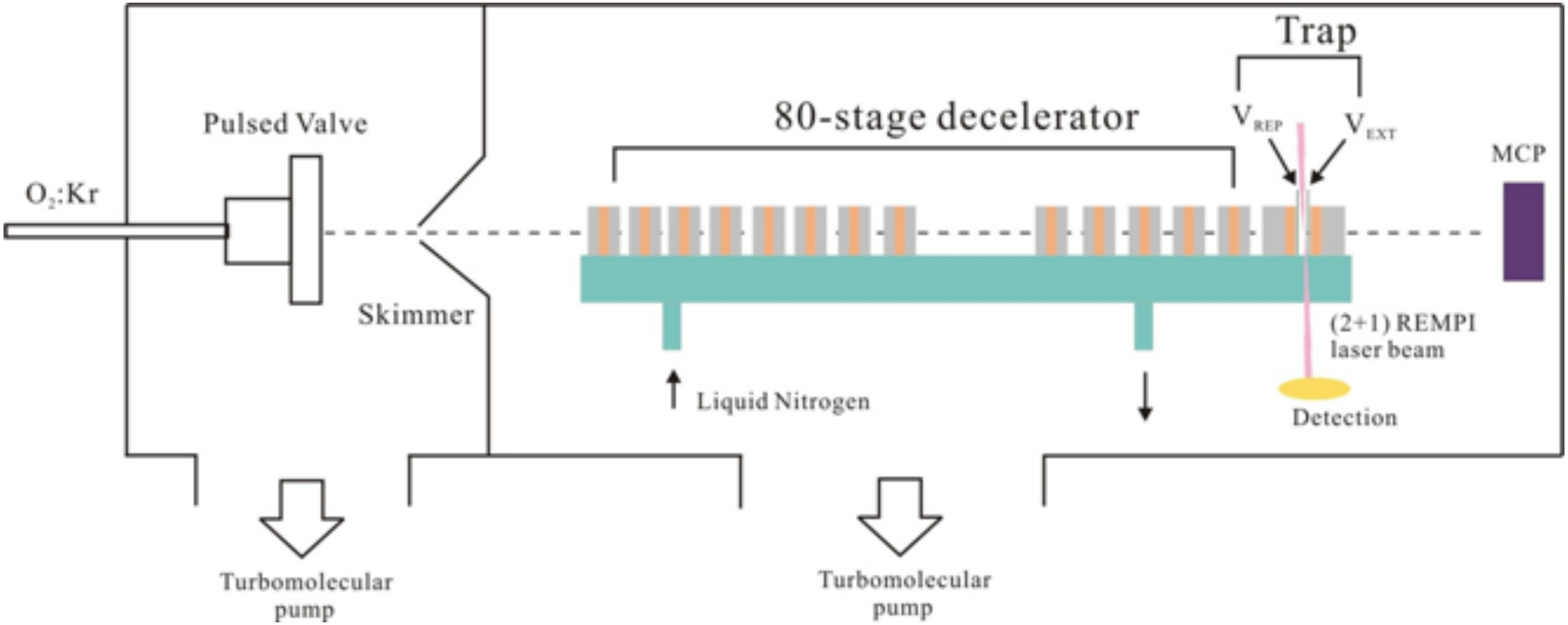






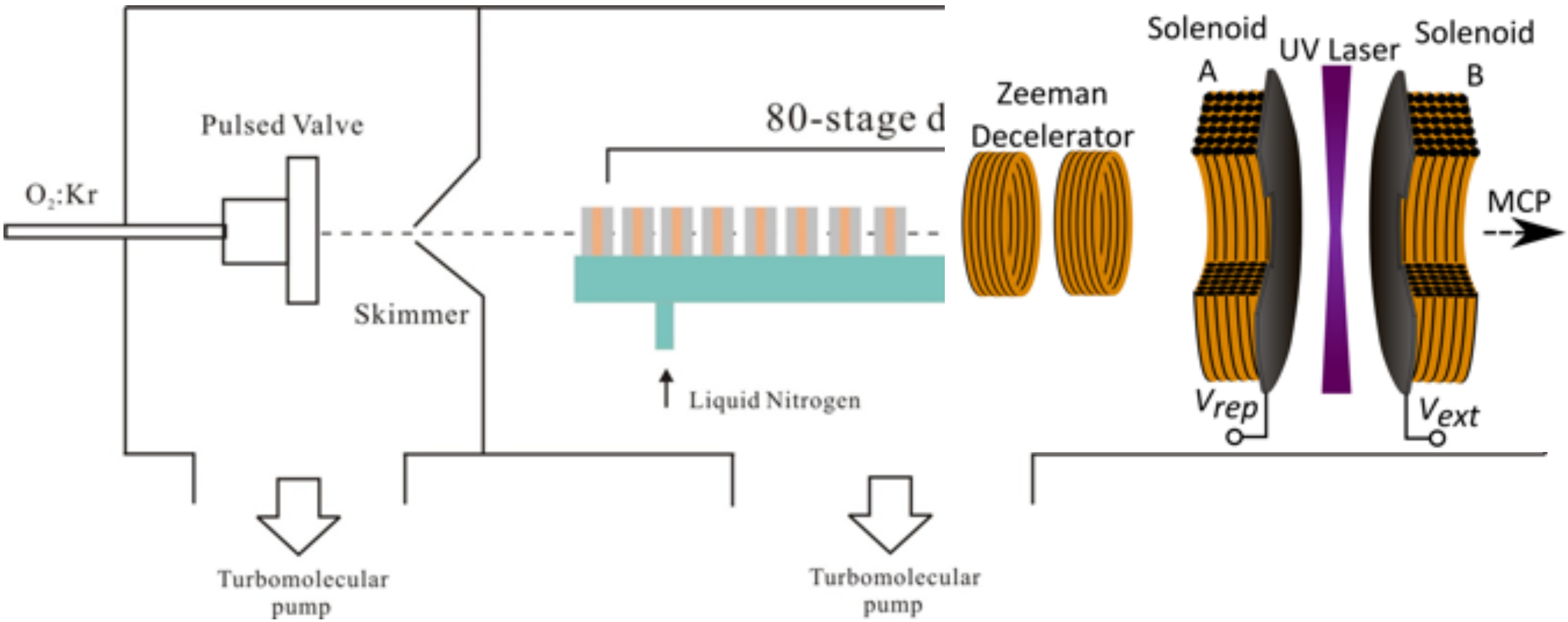
Trapping of Decelerated O_2

Anti-Helmholtz Coil



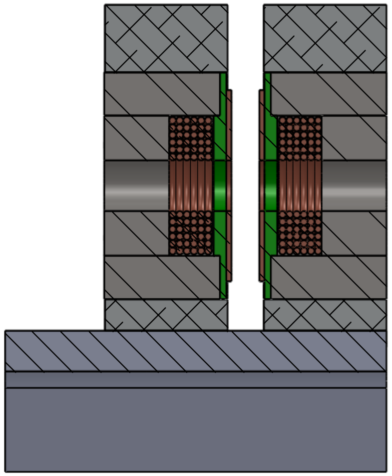
Trapping of Decelerated O_2

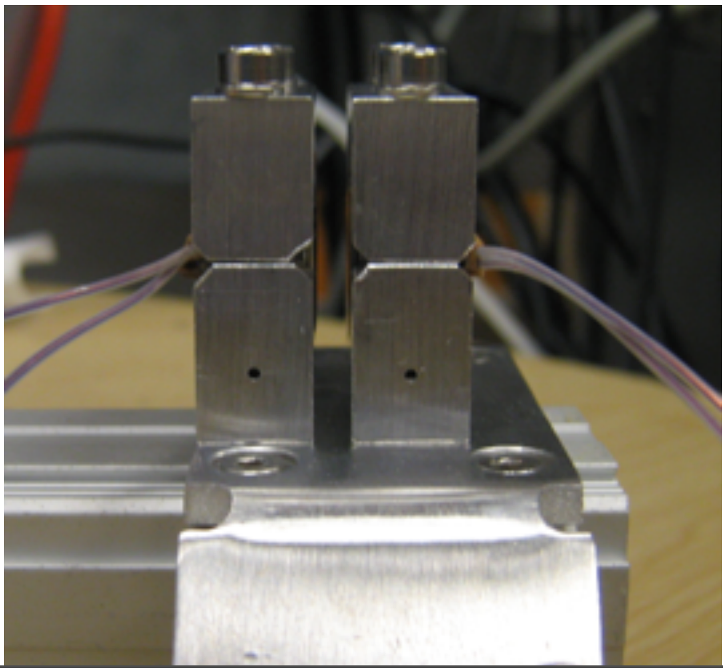
Anti-Helmholtz Coil



Trapping of O_2 in a Static Magnetic Field

anti-Helmholtz coil trap





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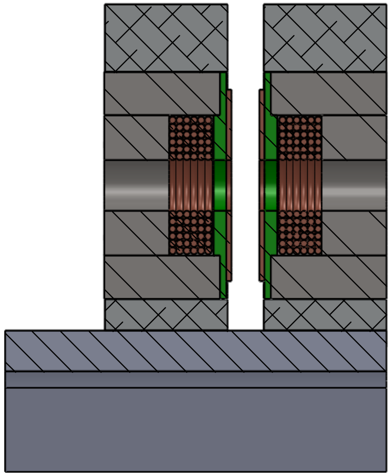
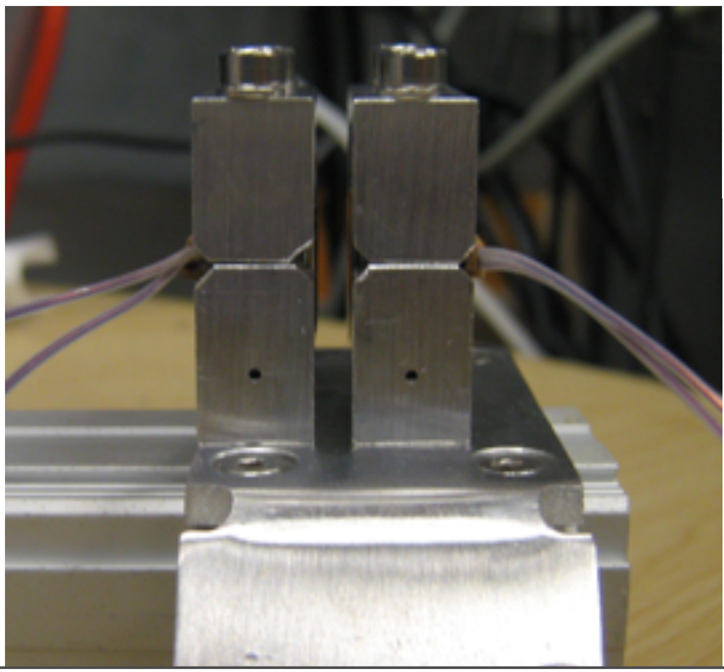
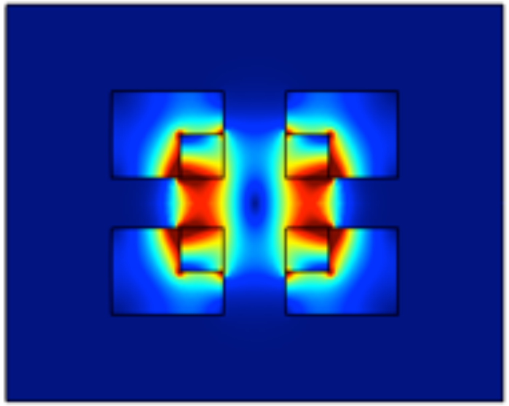
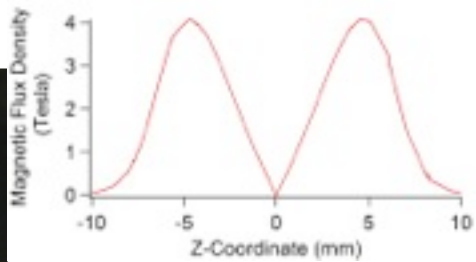




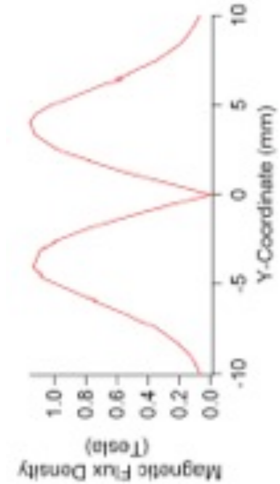
600 A





$E_{\perp} \sim 1T \sim 1.1cm^{-1}$

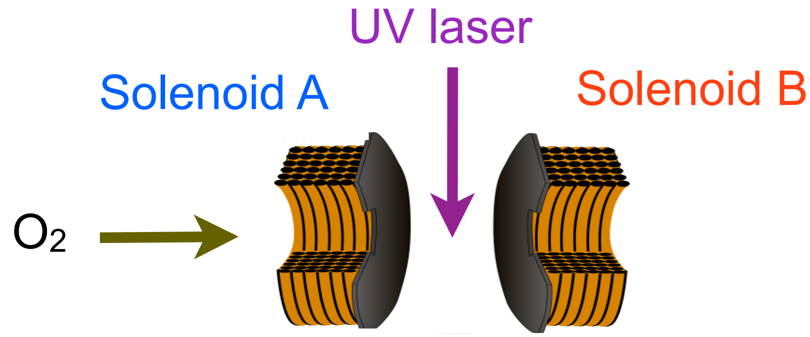
$\sim 28m/s\ (O_2)$



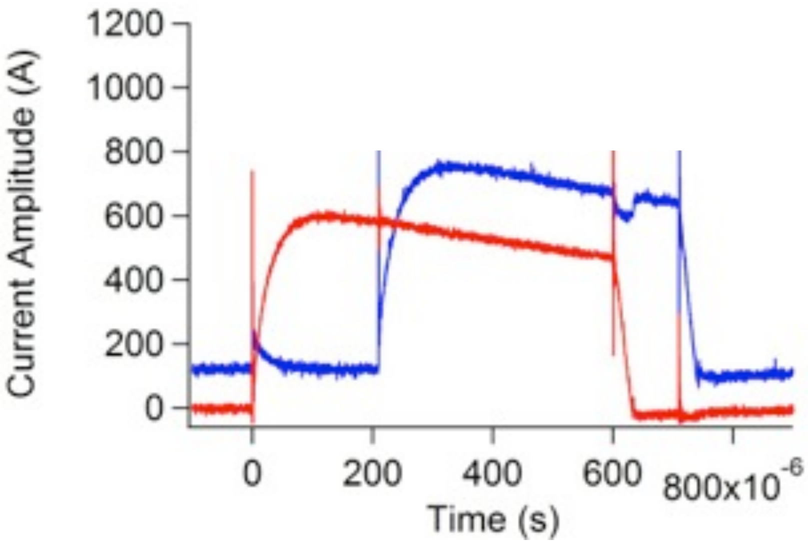
$E_{\parallel} \sim 4T \sim 3.6cm^{-1}$

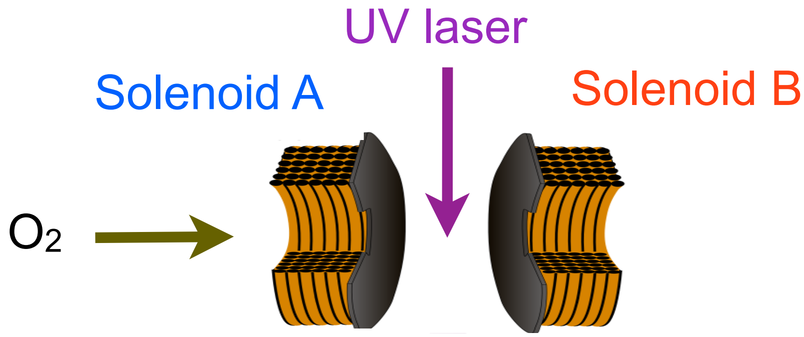
$\sim 52m/s\ (O_2)$

Trapping of O_2 in a Static Magnetic Field

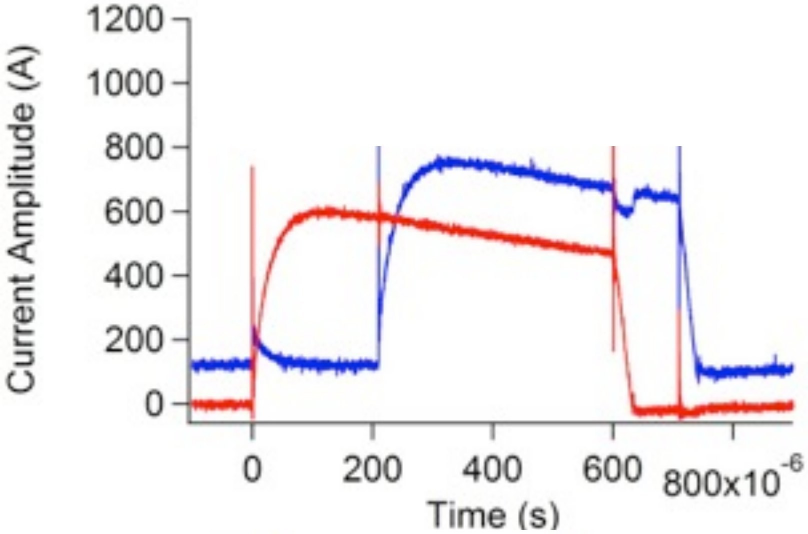


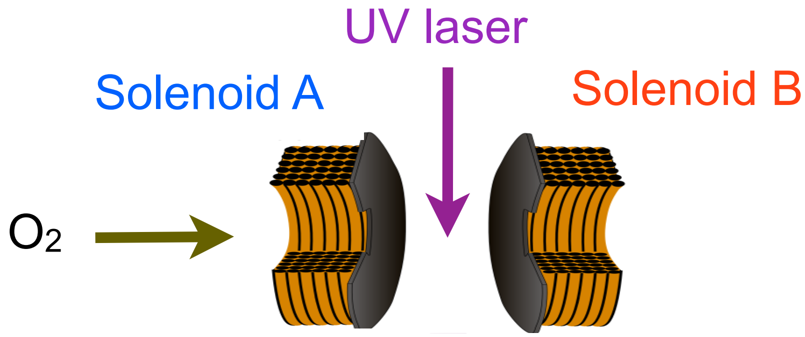
Trapping of O_2 in a Static Magnetic Field

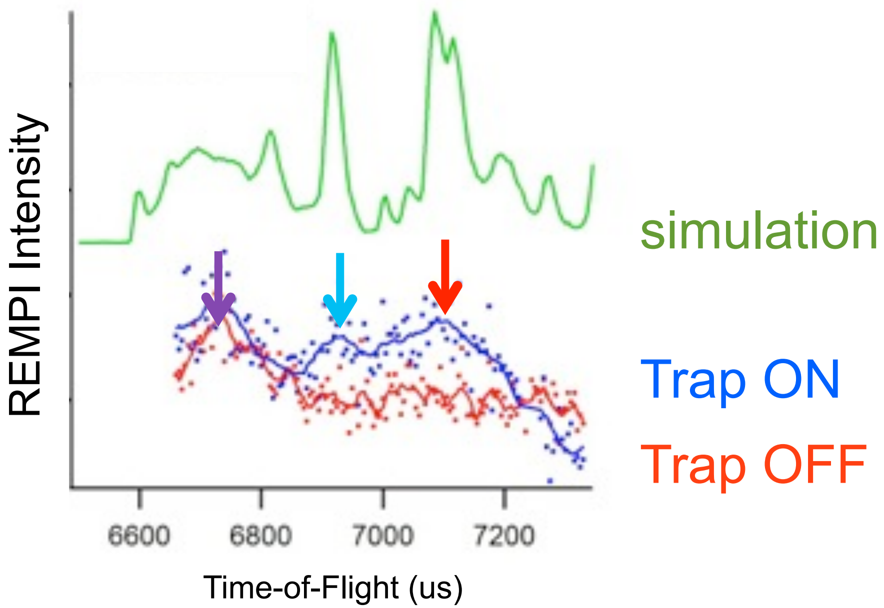




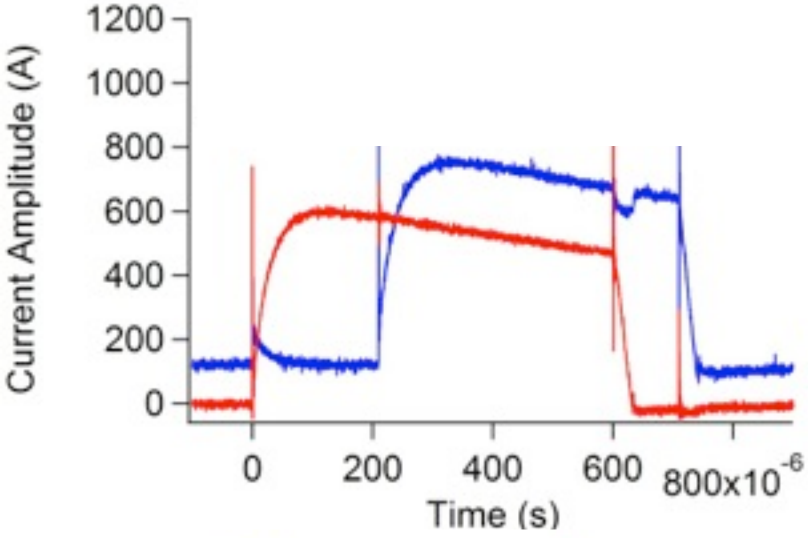
Trapping of O_2 in a Static Magnetic Field

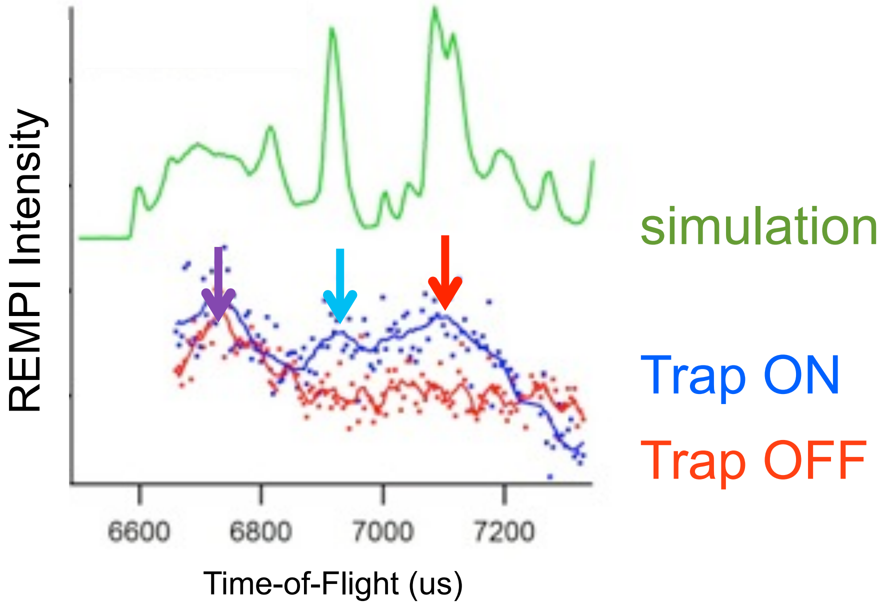


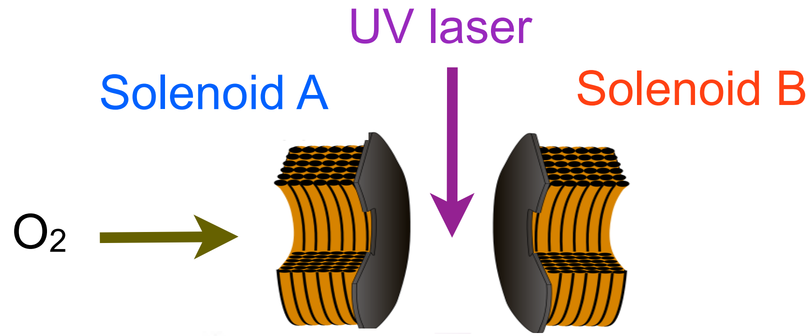


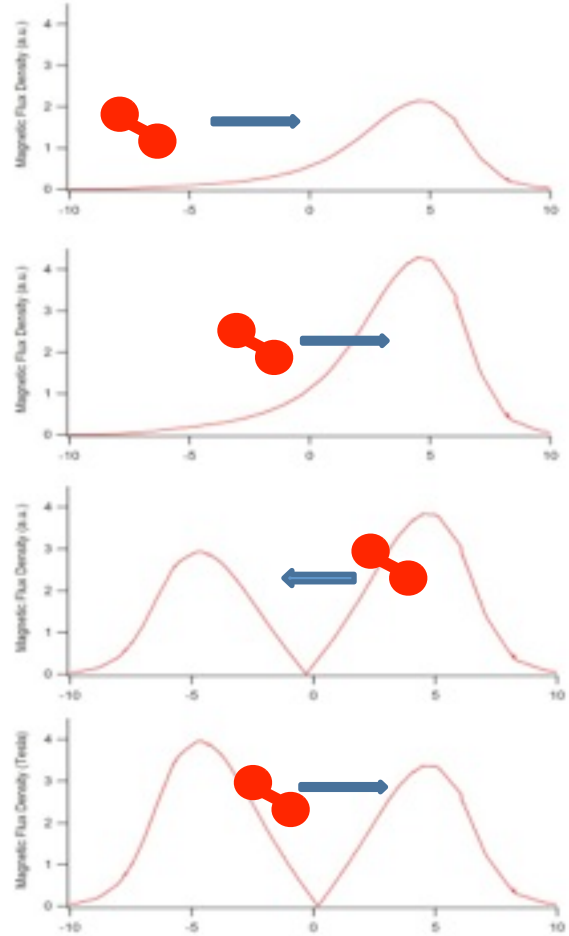


Trapping of O_2 in a Static Magnetic Field

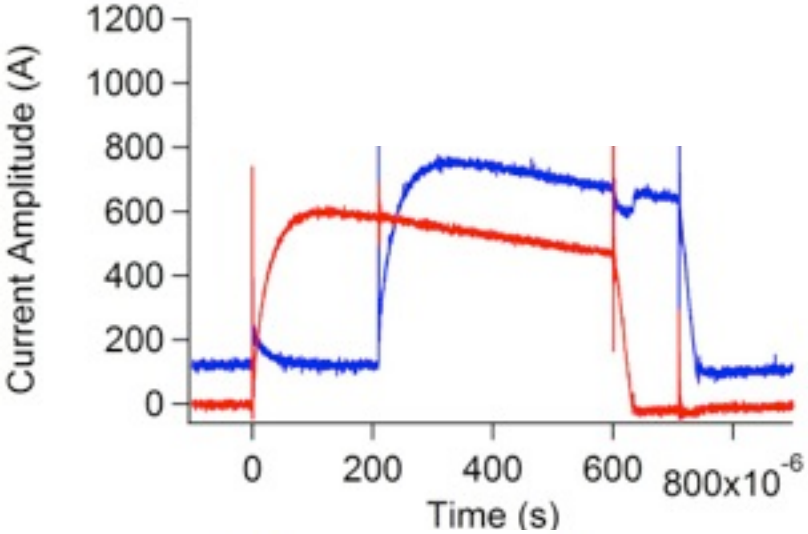


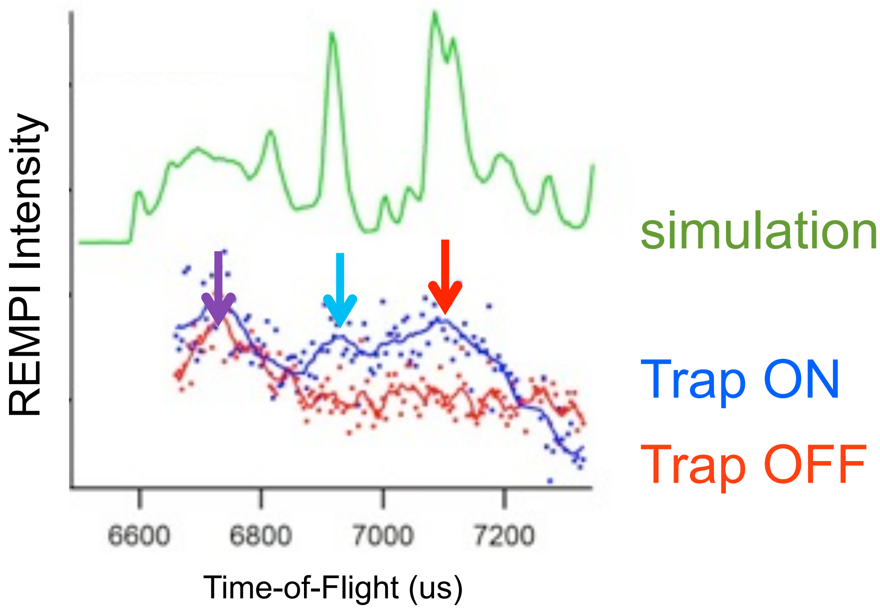




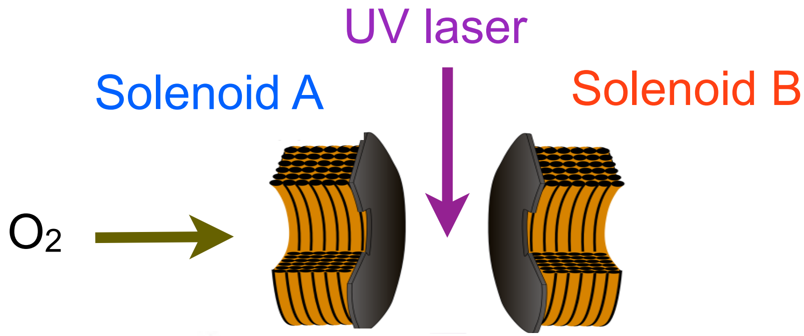


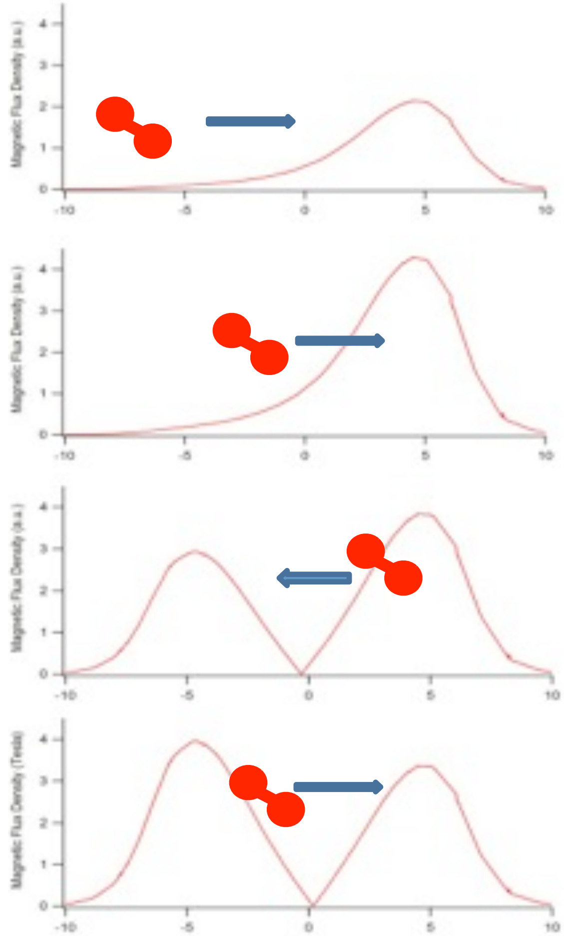
Trapping of O_2 in a Static Magnetic Field





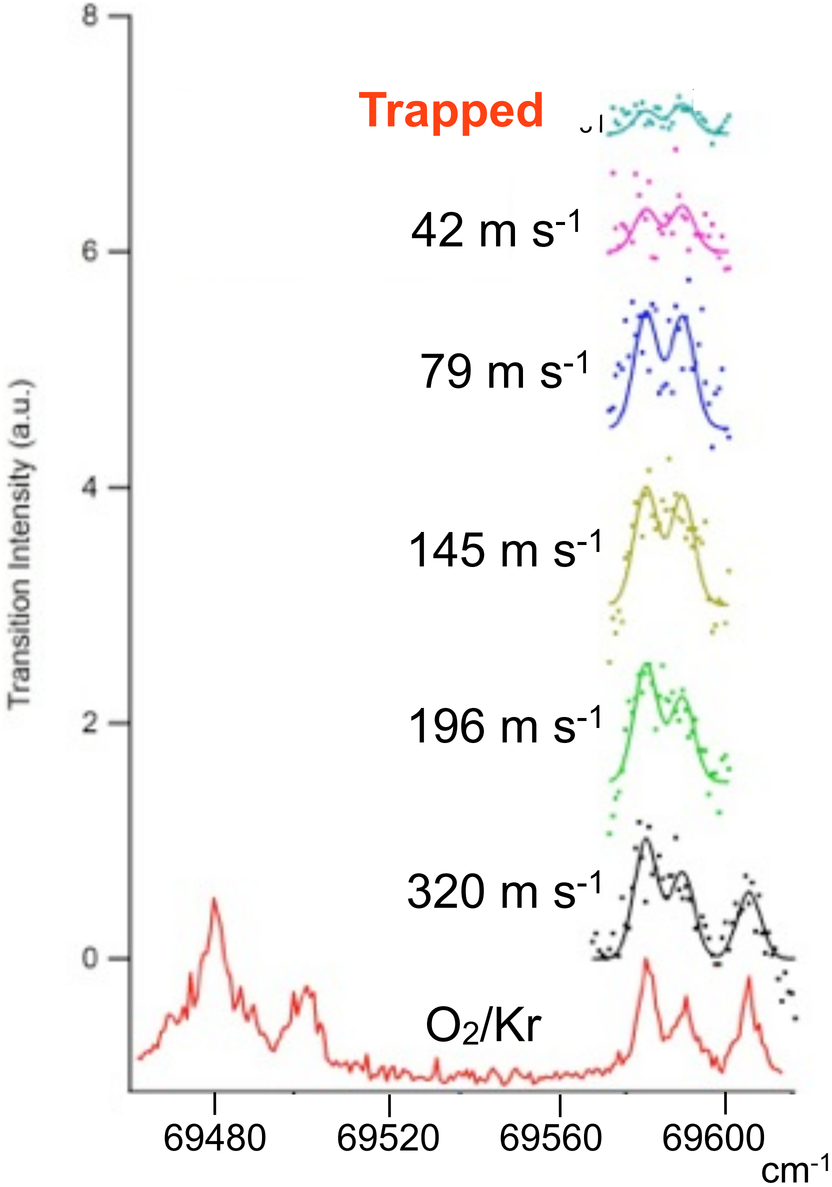
The first trapping signal of Zeeman decelerated molecules

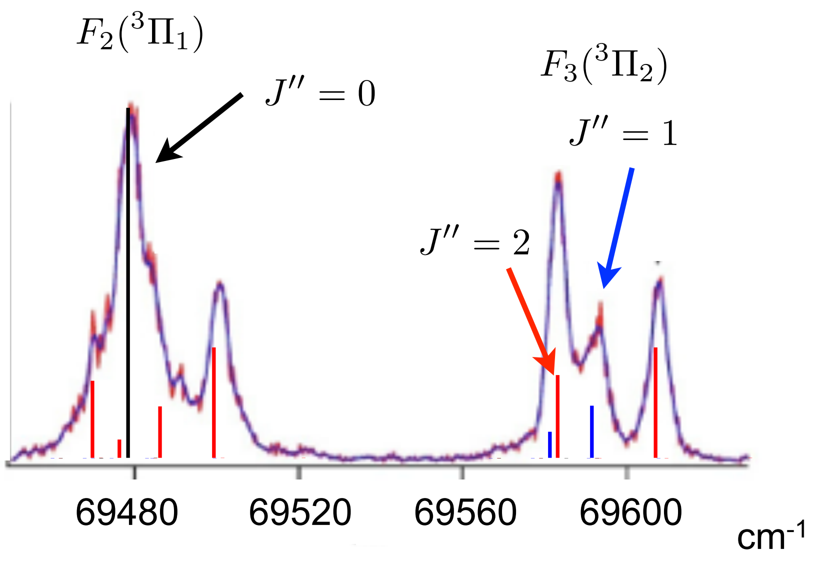


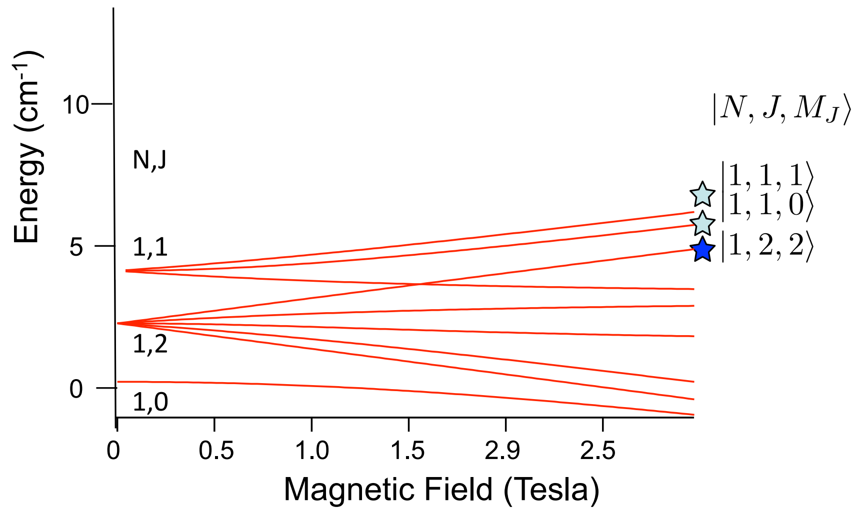


REMPI Spectra of Decelerated O_2

O2 in the |J=1, M_J=0,1> state are trapped



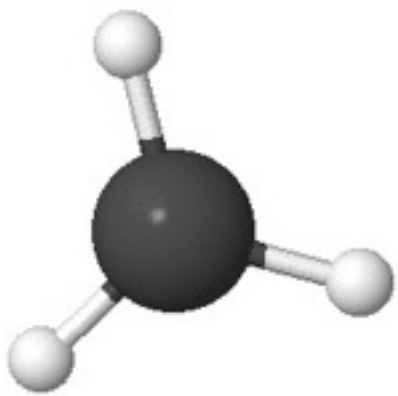




Deceleration of Free radicals

Deceleration of Free radicals

Methyl Radical: CH_3



CH_3

High-resolution Spectroscopy

A planer oblate top molecule

First Identification:

G. Herzberg and J. Shoosmith, Can. J. Phys. **34**, 523 (1956)

Matrix isolation spectroscopy:

D. E. Milligan and M. E. Jacox, J. Chem. Phys. **47**, 5146 (1967)

Flash Photolysis IR spectroscopy:

L. Y. Tan, A. M. Winer, and G. C. Pimentel, J. Chem. Phys. **57**, 4028 (1972).

ν_2 band high-resolution IR (606.45 cm^{-1}):

C. Yamada, E. Hirota, and K. Kawaguchi, J. Chem. Phys. **75**, 5256 (1981)

C. Yamada, and E. Hirota, J. Chem. Phys. **78**, 669 (1983)

μ_{10}=0.28 D

ν_3 band high-resolution IR (3160.82 cm^{-1}):

T. Amano, P. Bernath, C. Yamada, Y. Endo, and E. Hirota, J. Chem. Phys. **77**, 5284 (1982)

μ_{10}=0.1 D

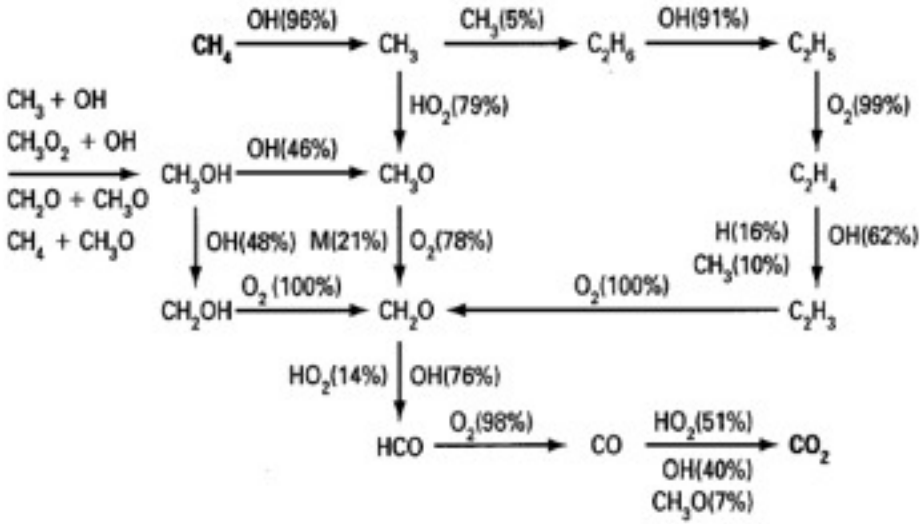
(slit jet) S. Davis, D.T. Anderson, G Duxbury, and D. J. Nesbitt, J. Chem. Phys. **107**, 5661 (1997)

Detected in Space

H. Feuchtgruber, F.P. Helmich, E.F. van Dishoeck and C.M. Wright, Astro. J. **535**, L111 (2000).

CH_3

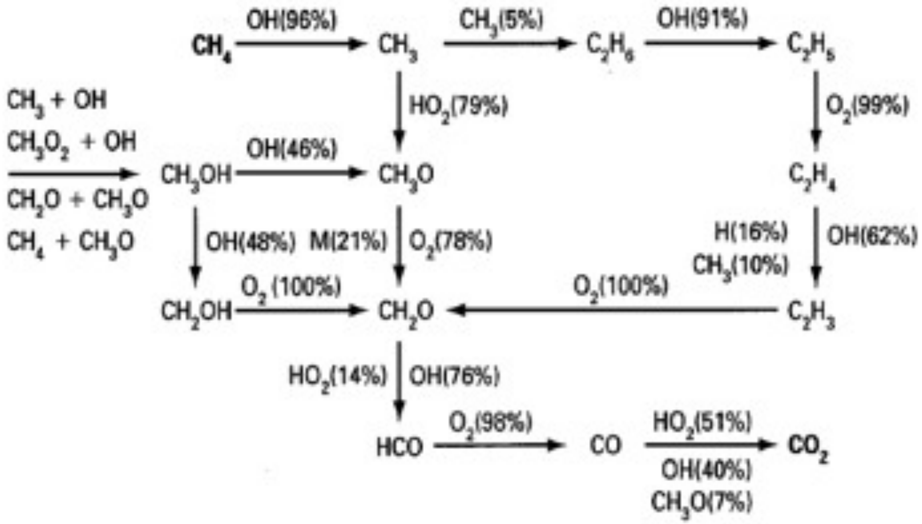
Reactive Species



Methane oxidation mechanism

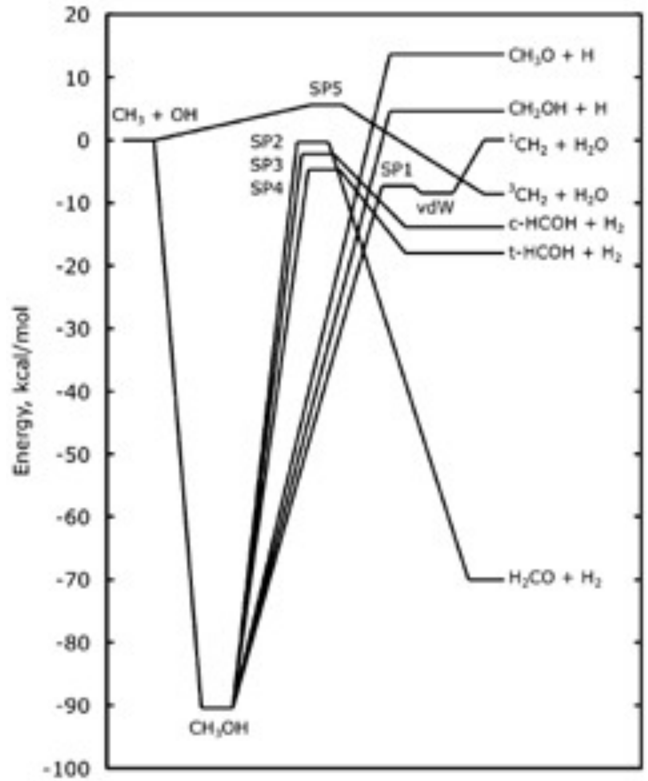
CH_3

Reactive Species



Methane oxidation mechanism

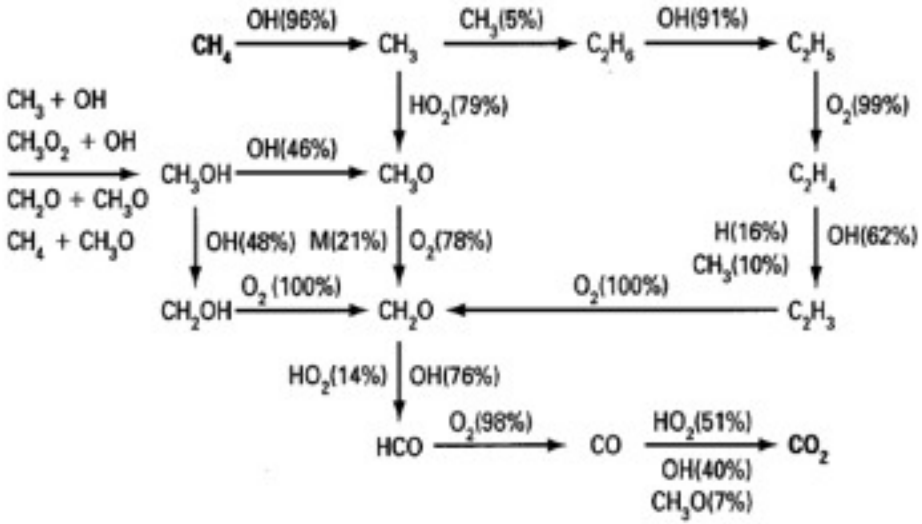
CH_3 + OH Barrierless reaction



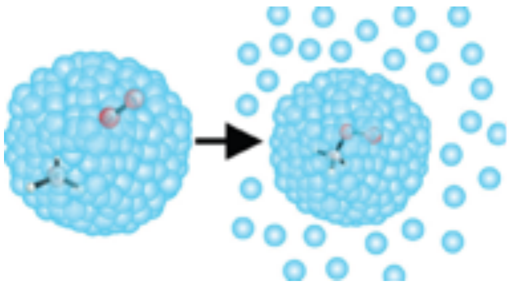
Ahren W. Jasper et al,
J.Phys.Chem. A **111**, 3932(2007)

CH_3

Reactive Species



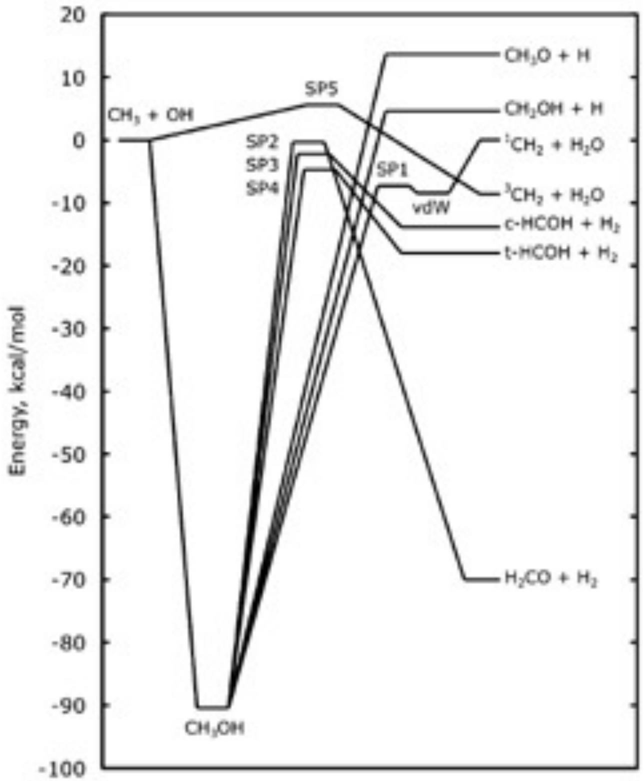
Methane oxidation mechanism



$CH_3 + O_2 \rightarrow CH_3OO$ at 0.4K

A. M. Morrison et al,
J.Phys.Chem.A **116**, 5299(2012)

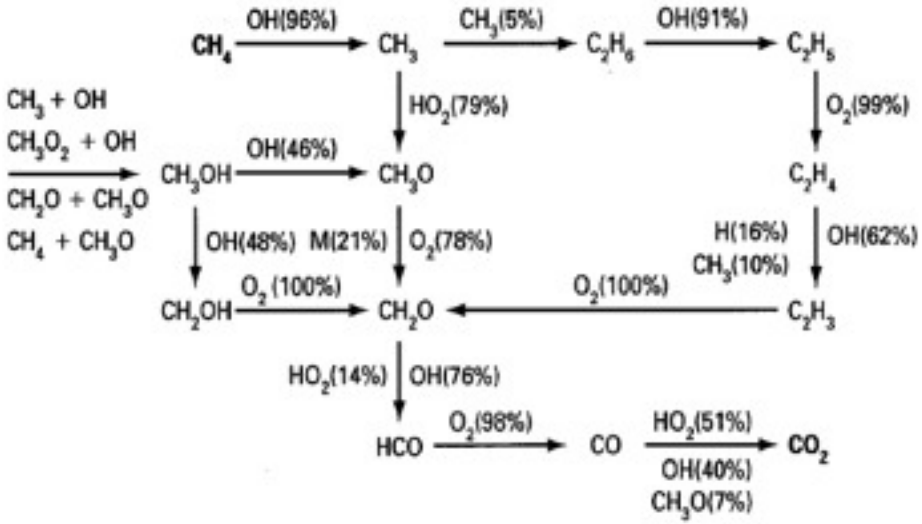
CH_3 + OH Barrierless reaction



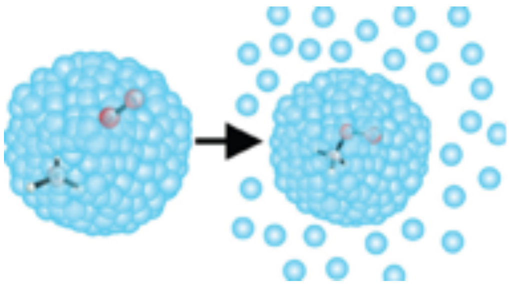
Ahren W. Jasper et al,
J.Phys.Chem. A **111**, 3932(2007)

CH_3

Reactive Species



Methane oxidation mechanism



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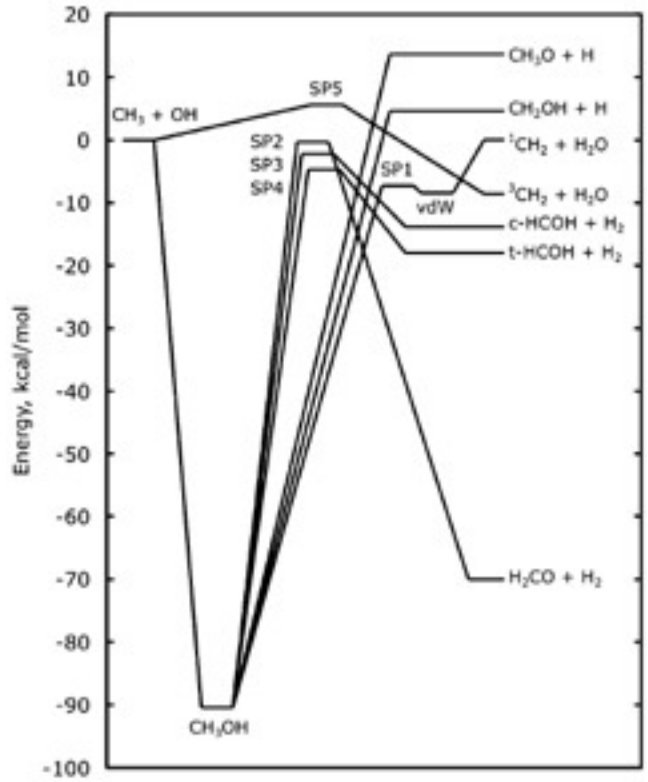
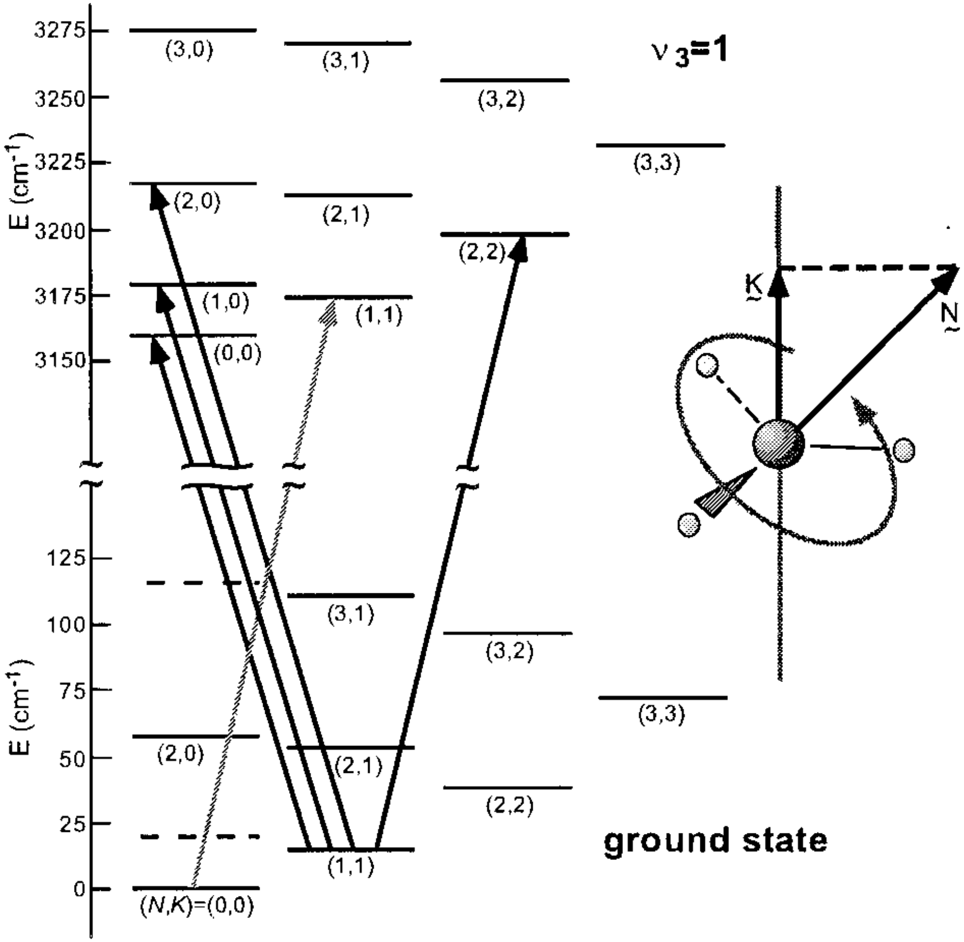


Ahren W. Jasper et al, J.Phys.Chem. A **111**, 3932(2007)

Cold chemistry !

CH$_3$

ortho-para modification



Davis et al. J. Chem. Phys. 107, 5661 (1997)

study of ortho-para conversion

Closed cycle IR transitions

IR laser cooling

Sympathetic cooling with Li/Rb

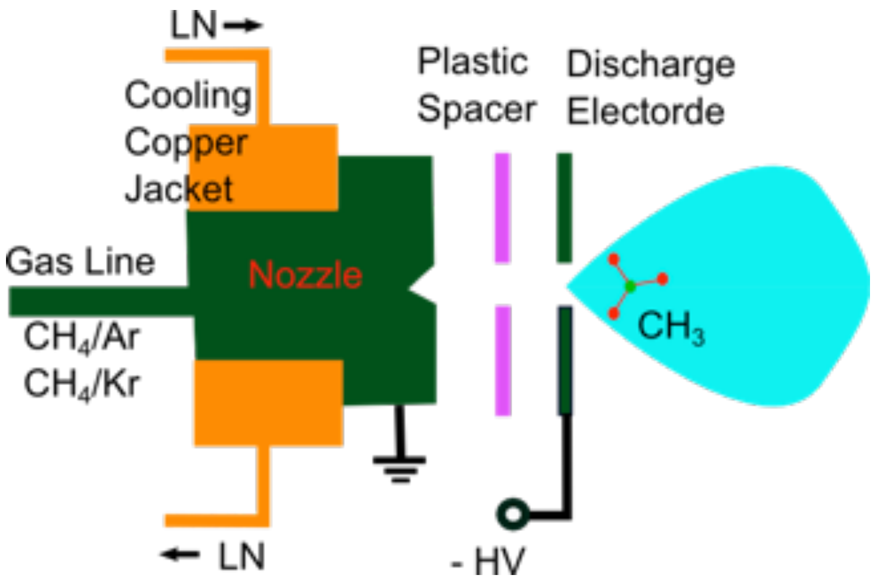
T.V. Tscherbul et al.
Phys. Rev. Lett. **106**, 073201 (2011).

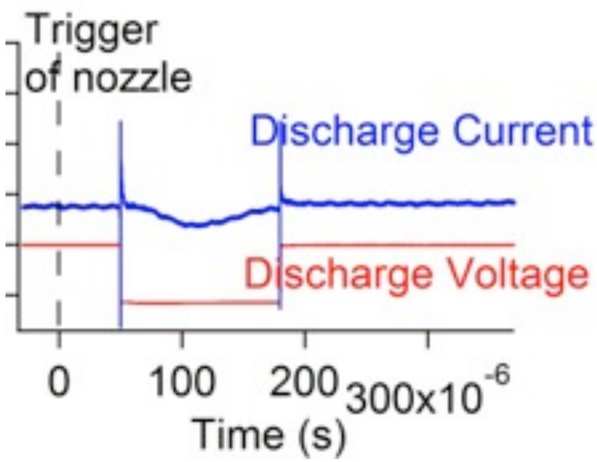
Various possibilities to cool below 1 mK

Production of CH_3

CRUCS valve

Liq. N_2 cooling
Opening time : 30~60us
4- 6 bar pressure





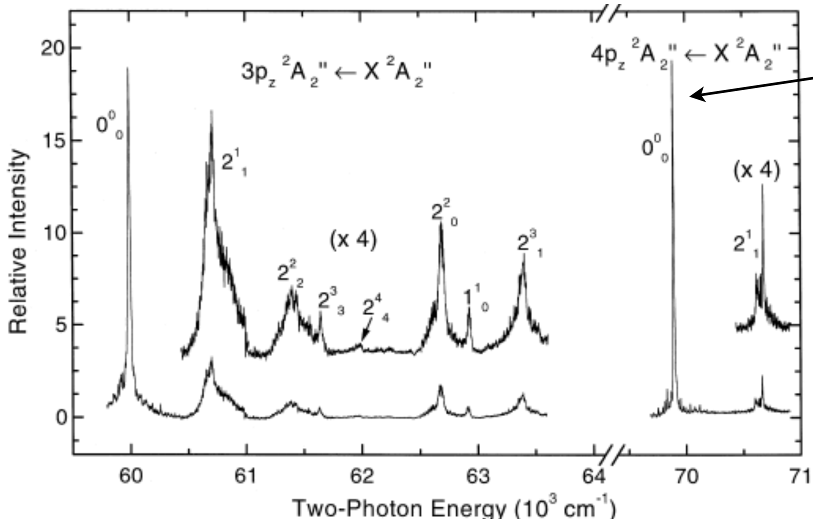
Discharge period: 150us
FWHM beam width 50 us

A compact radical beam

CH_3 Detection : 2+1 REMPI

B. Martínez-Haya, et al. Chem. Phys. Lett. 311, 159 (1999)

$$4p\ {}^2A_2'' \leftarrow \tilde{X}\ {}^2A_2'' \quad 0_0^0$$



Rydberg transition

resonant at 4×17,470cm^{-1}

O_2 at 4×17,400cm^{-1}

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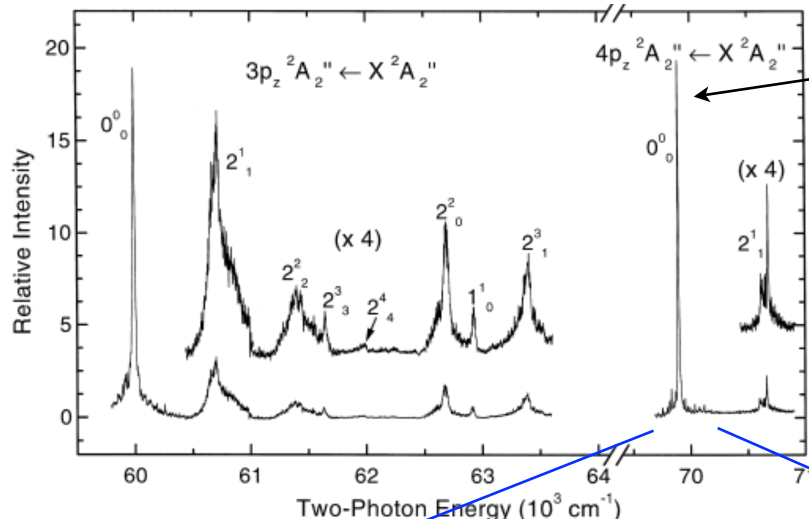
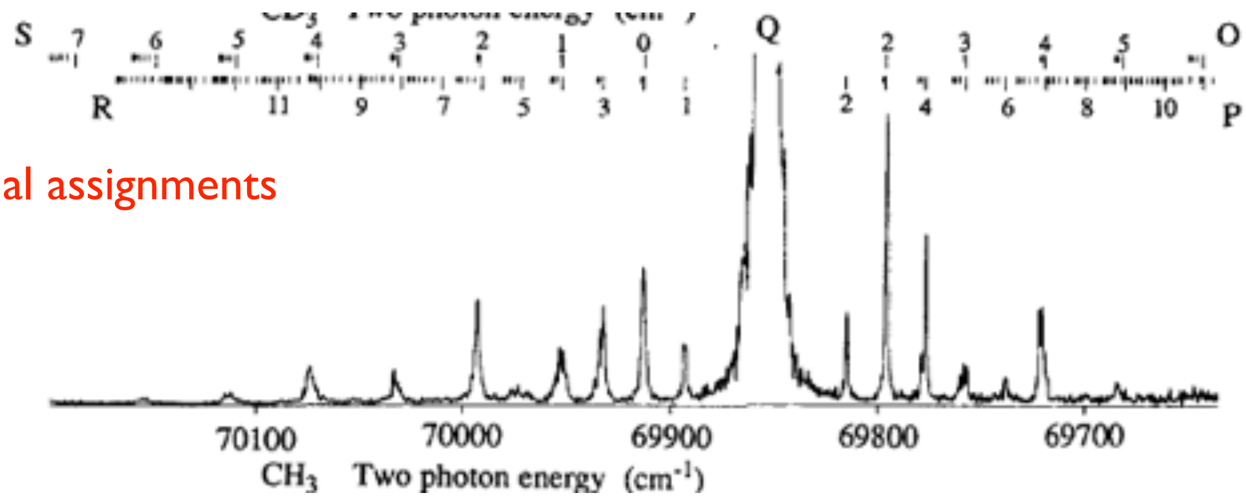


resonant at 4×17,470cm^{-1}

O_2 at 4×17,400cm^{-1}

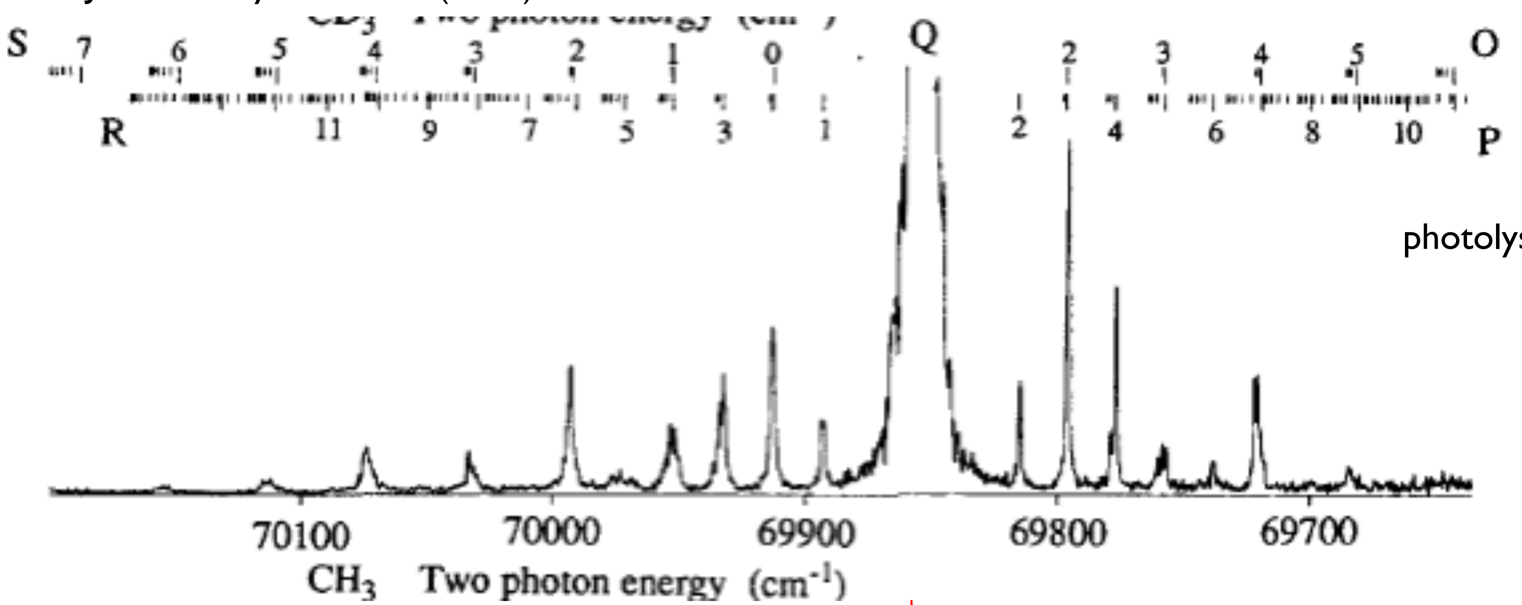
Black, Powis J. Chem. Phys. 89, 3986 (1988)

rotational assignments



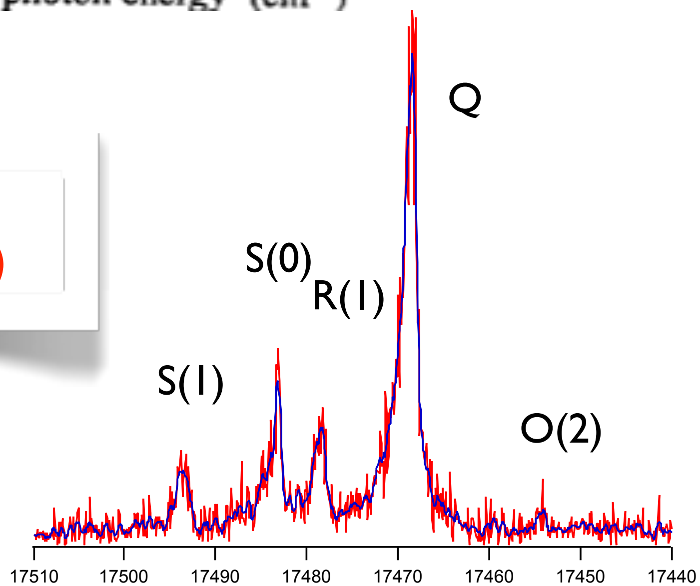
CH_3 Radical Beam

Black, Powis J. Chem. Phys. 89, 3986 (1988)



photolysis of CH_3I

Discharge of CH_4/Ar with a cold nozzle (150K)



rotational temp <5 K

N"=0 : N"=1 2:1

CH_3 Radical Beam

Detection is 1 m downstream of a nozzle.

Requirements for the Zeeman deceleration:

High-density, low rotational temperature, well collimated, slow, and background free radical beam

CH_3 Radical Beam

Detection is 1 m downstream of a nozzle.

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UV dissociation of CH_3I

CH_3 Radical Beam

Detection is 1 m downstream of a nozzle.

Requirements for the Zeeman deceleration:

High-density, low rotational temperature, well collimated, slow, and background free radical beam

UV dissociation of CH_3I

UV dissociation or discharge of Di-tert-butyl peroxide

CH_3 Radical Beam

Detection is 1 m downstream of a nozzle.

Requirements for the Zeeman deceleration:

High-density, low rotational temperature, well collimated, slow, and background free radical beam

UV dissociation of CH_3I strong background REMPI signal

UV dissociation or discharge of Di-tert-butyl peroxide

CH_3 Radical Beam

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UV dissociation or discharge of Di-tert-butyl peroxide

high rotational temperature

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UV ~~dissociation or discharge~~ of Di-tert-butyl peroxide

high rotational temperature

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Detection is 1 m downstream of a nozzle.

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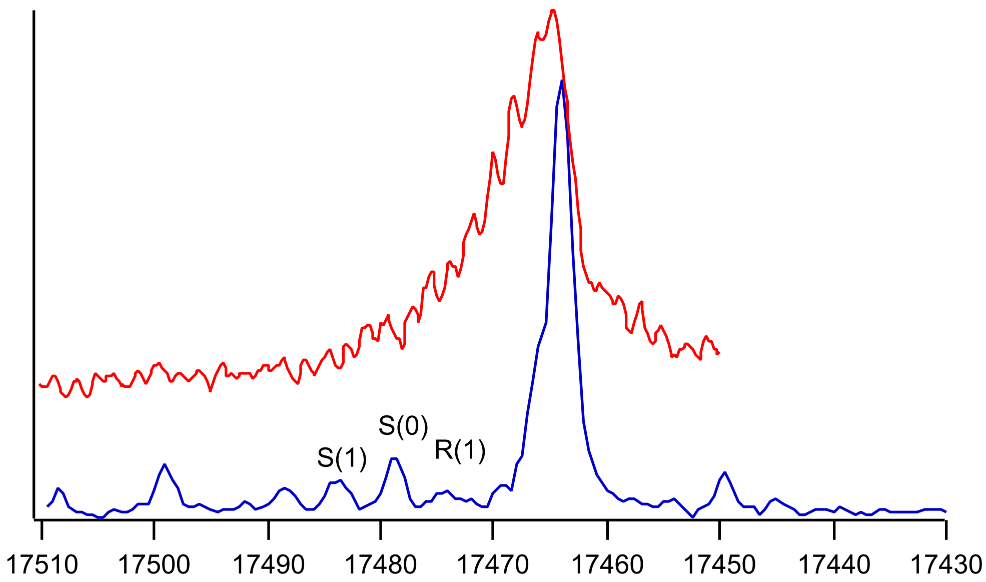
~~UV dissociation of CH_3I~~ strong background REMPI signal

~~UV dissociation or discharge of Di-tert-butyl peroxide~~

high rotational temperature

peroxide vs CH_4 discharge



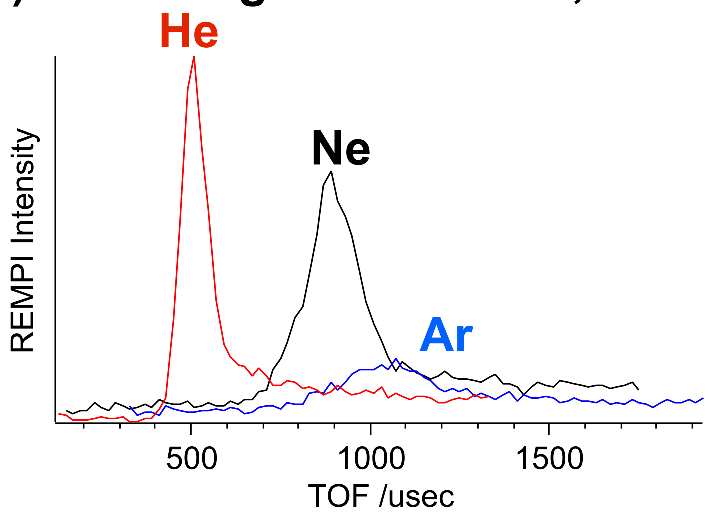
Production of CH_3

Discharge of CH_4/Ar

Production of CH_3

Discharge of CH_4/Ar

(1) He seed gas is the best, but too fast.



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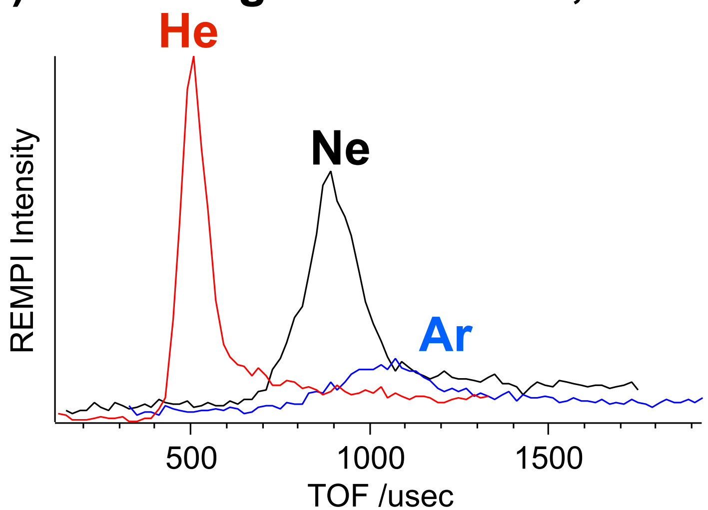
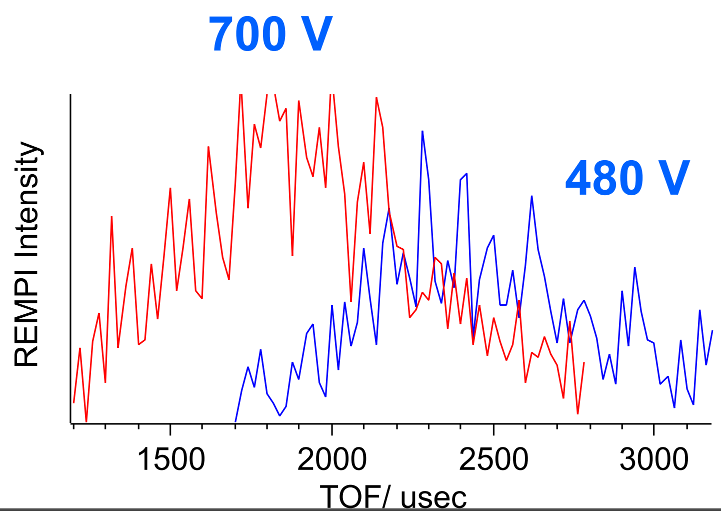


(2) High-discharge voltage causes acceleration of radical beams



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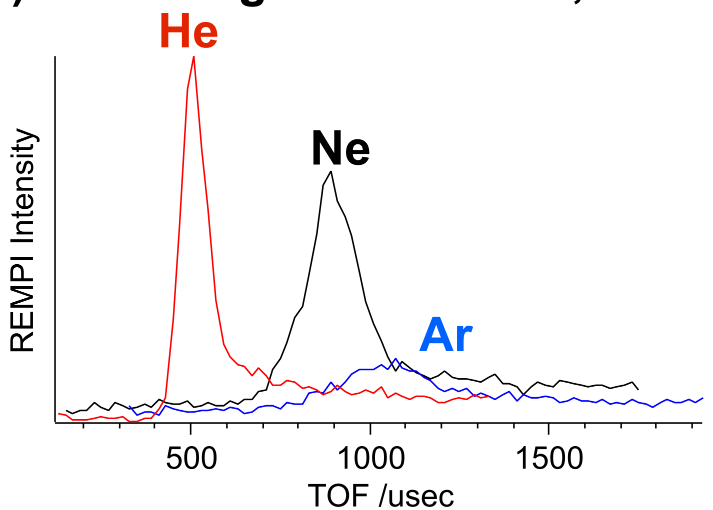

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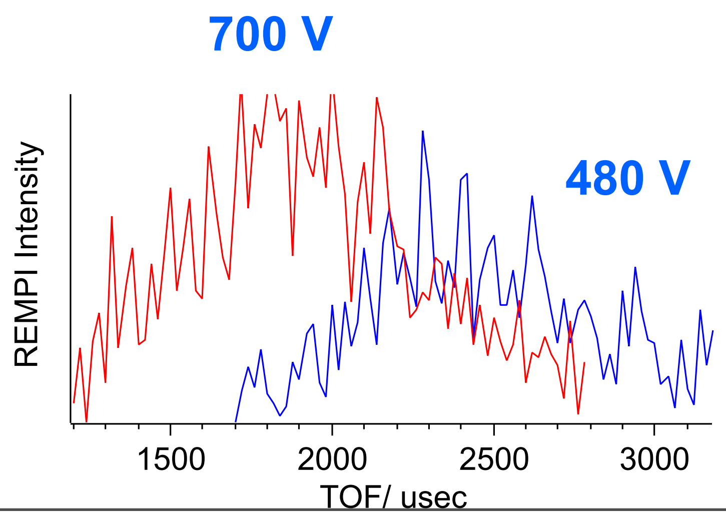
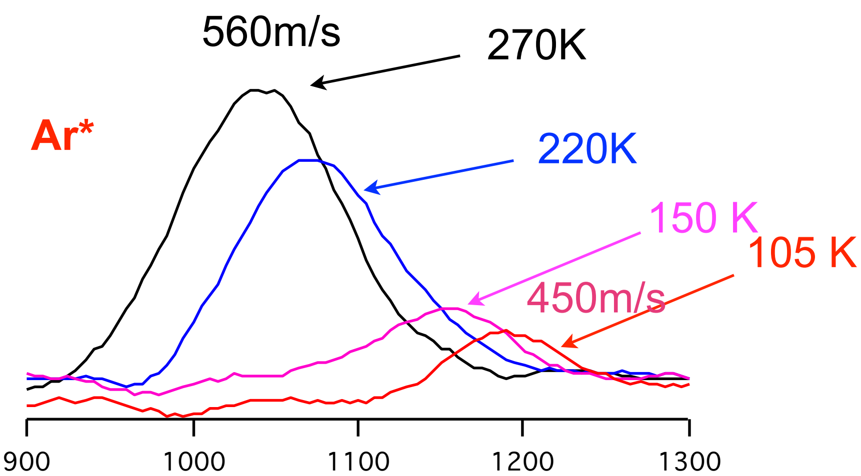
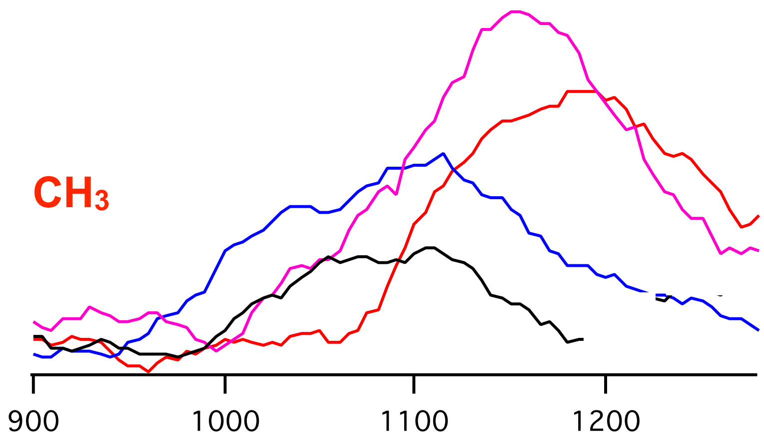


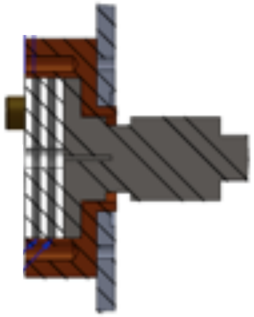
(3) Cooling nozzle to 150 K makes the beam slow and intense



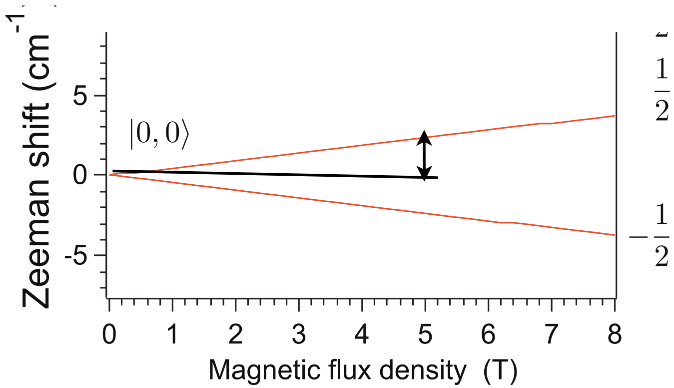


CH_3 signals are most intense at 150 K

CH_4/Ar discharge



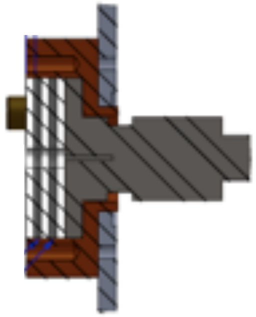
1.25 cm^{-1} /coil



Phys. Chem. Chem. Phys **15**, 1772 (2013)

Deceleration of CH_3

CH_4/Ar discharge



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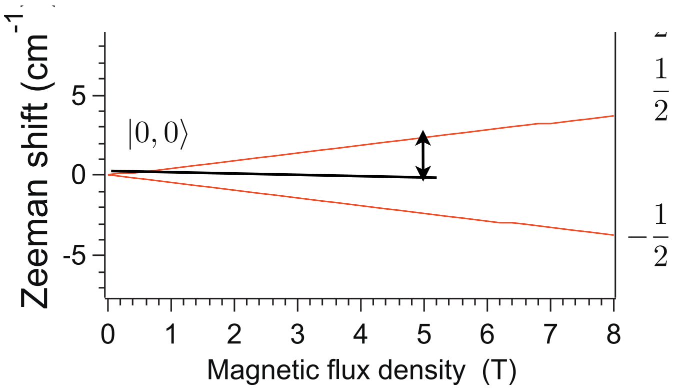
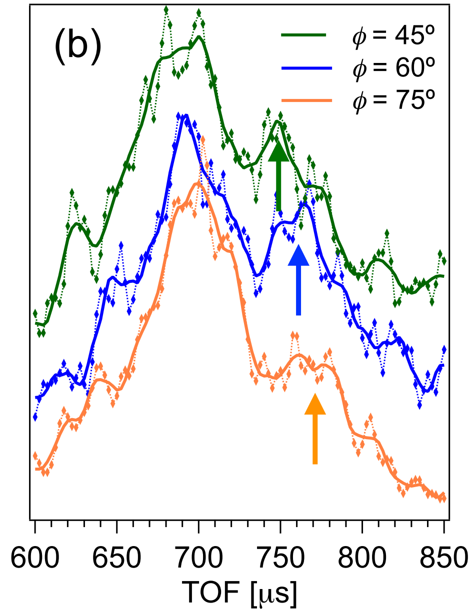
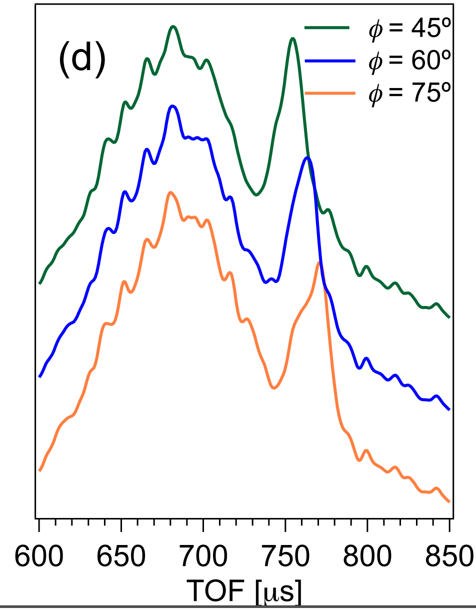


15-coil system



expt.

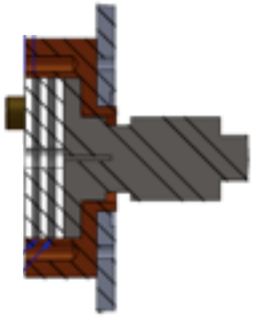


sim.

Phys. Chem. Chem. Phys **15**, 1772 (2013)

Deceleration of CH_3

CH_4/Ar discharge



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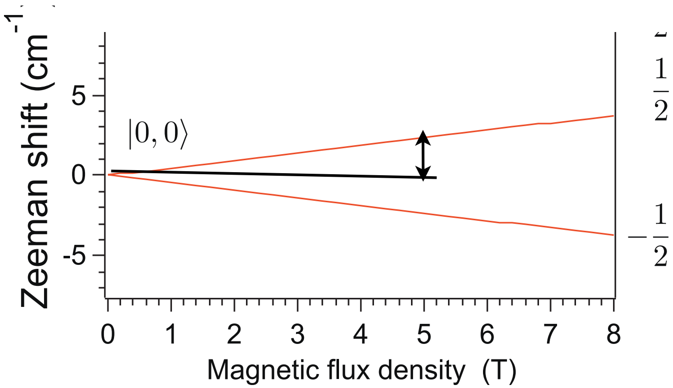
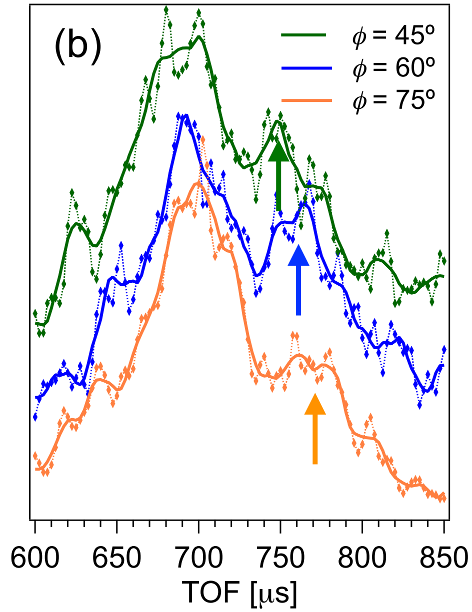


510 m/s → 470 m/s

ΔE ~ −19 cm^{-1} ~ 28 K

Phys. Chem. Chem. Phys **15**, 1772 (2013)

15-coil system



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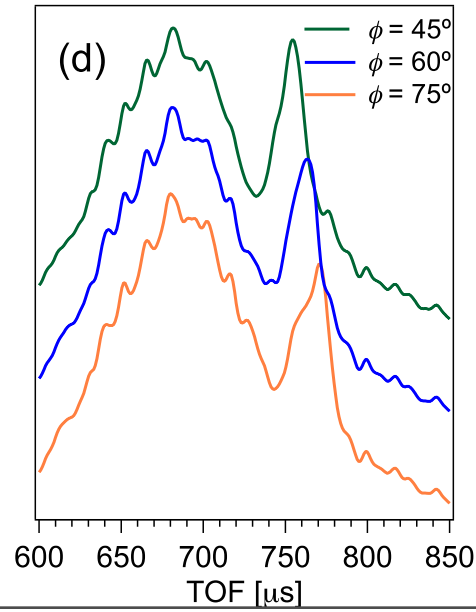
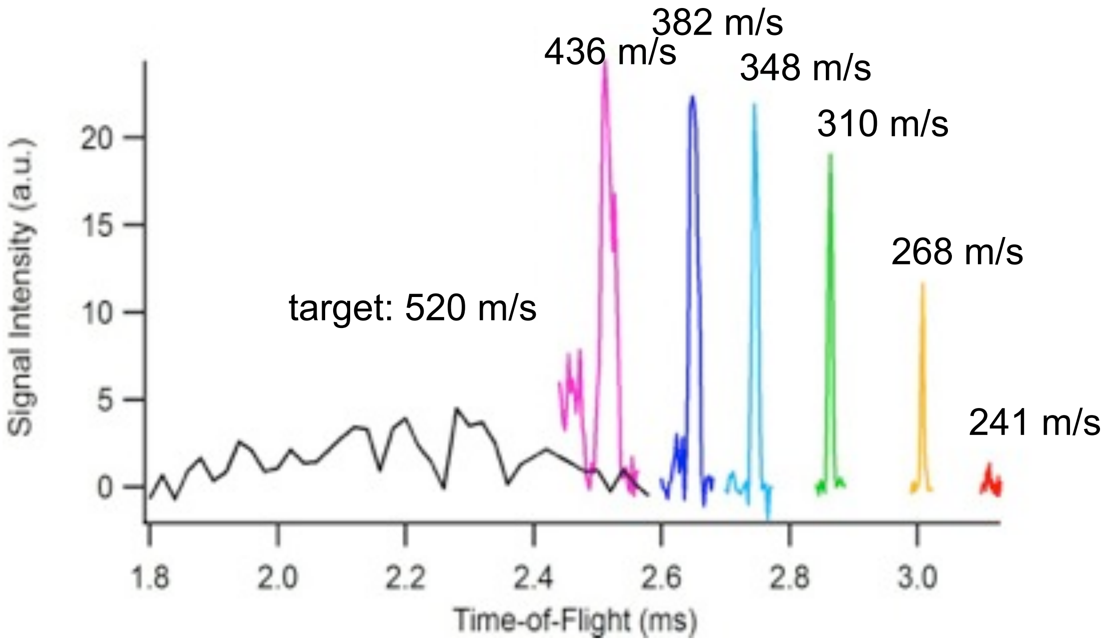


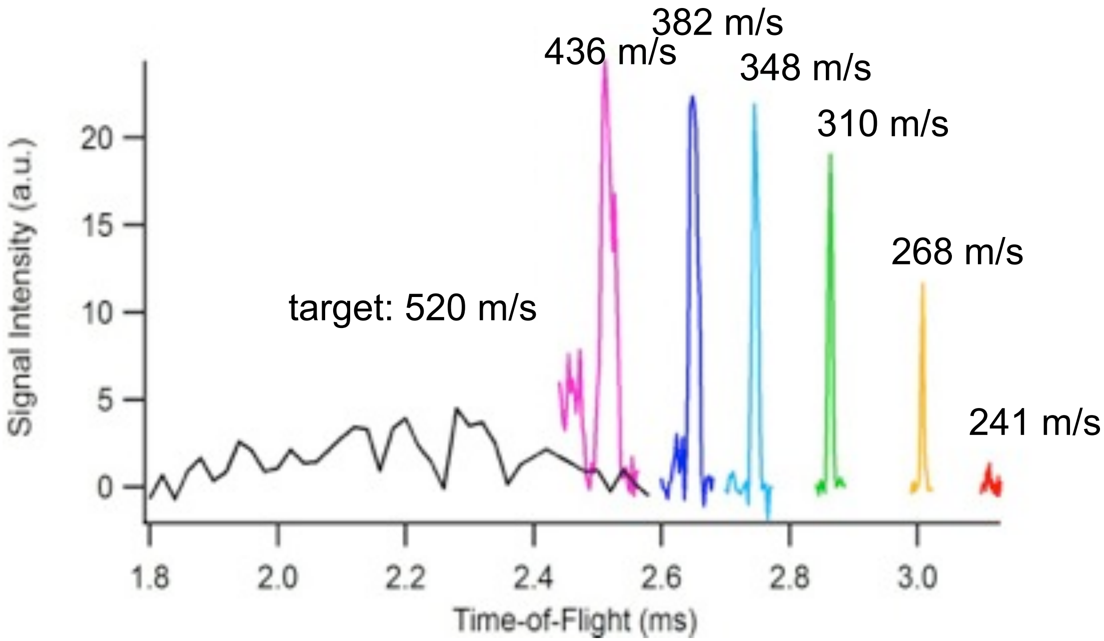
sim.

Deceleration of CH_3 : 80 Coil System



Sample: 6.2bar 20%CH4/Ar
Nozzle: 140K

Deceleration of CH_3 : 80 Coil System

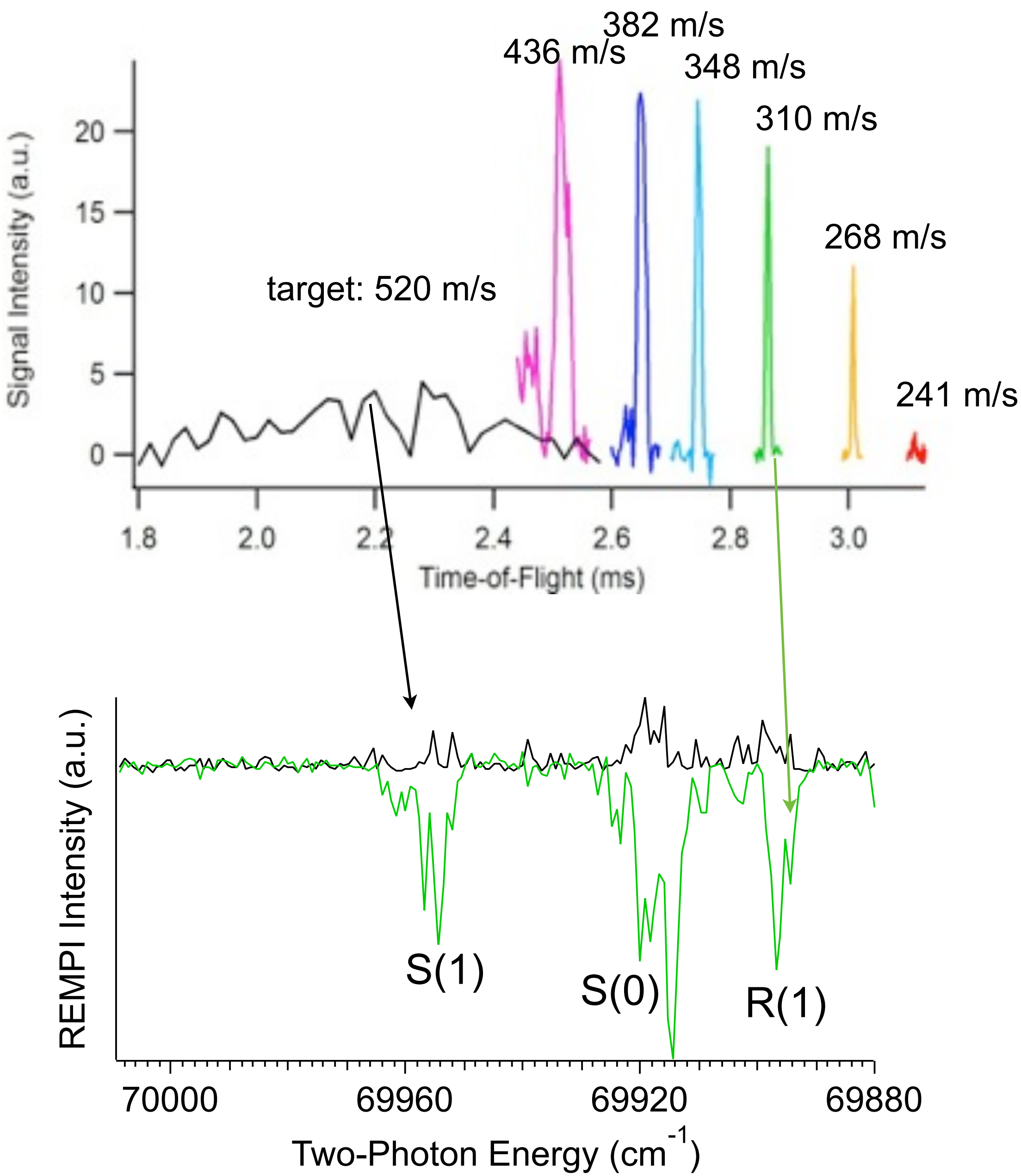


Sample: 6.2bar 20%CH4/Ar
Nozzle: 140K

Strong transverse focusing

CH_3 beam is less collimated

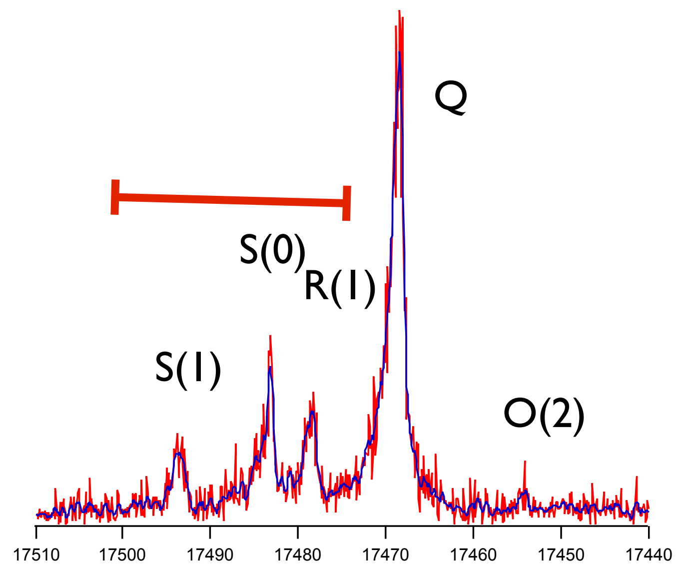
Deceleration of CH_3 : 80 Coil System



Sample: 6.2bar 20%CH4/Ar
Nozzle: 140K

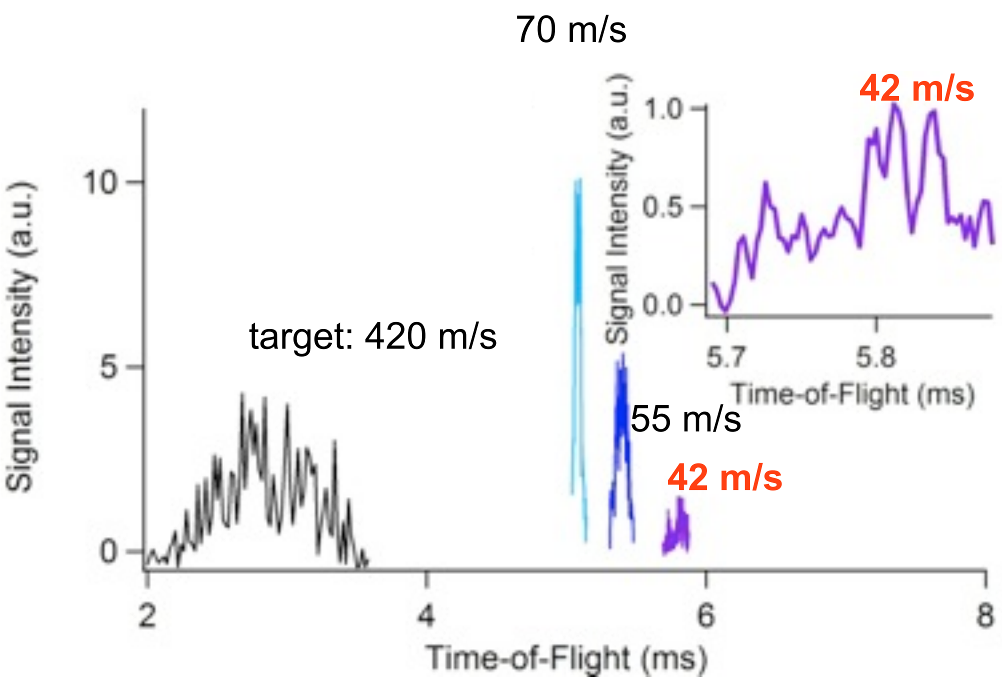
Strong transverse focusing

CH_3 beam is less collimated



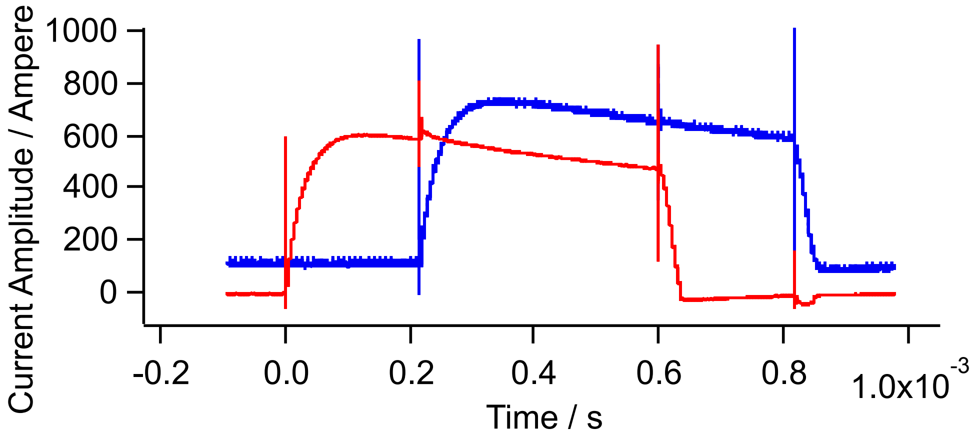
Both *N* = 0 and 1 states are decelerated

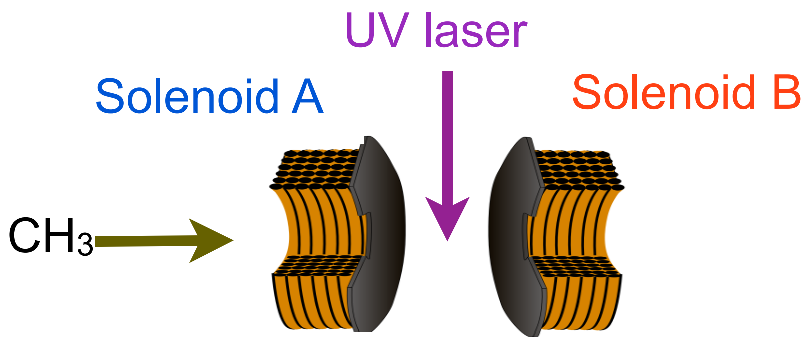
Deceleration of CH_3 : 80 Coil System

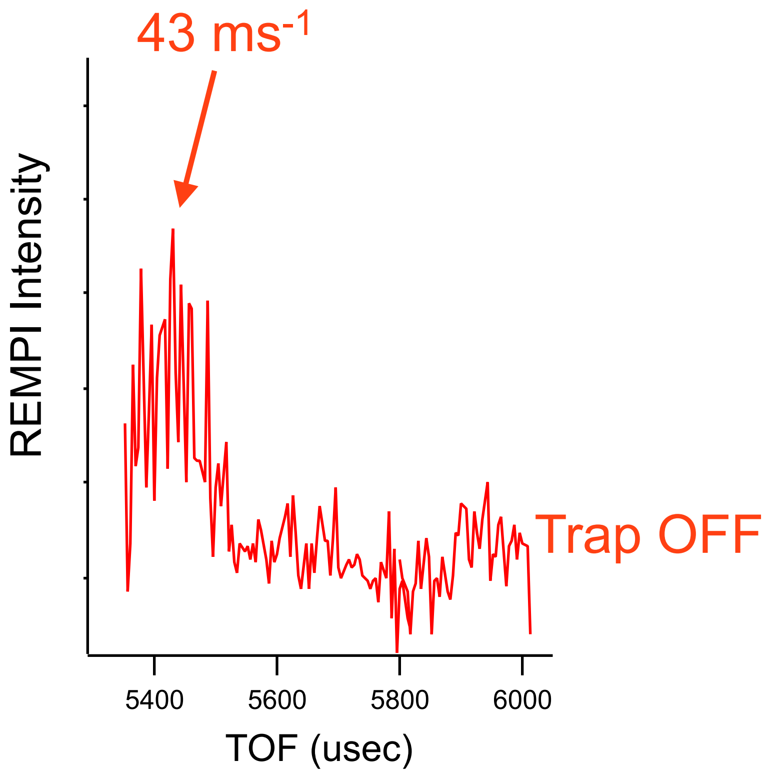


Sample: 5bar 30%CH4/Kr
Nozzle: 150K

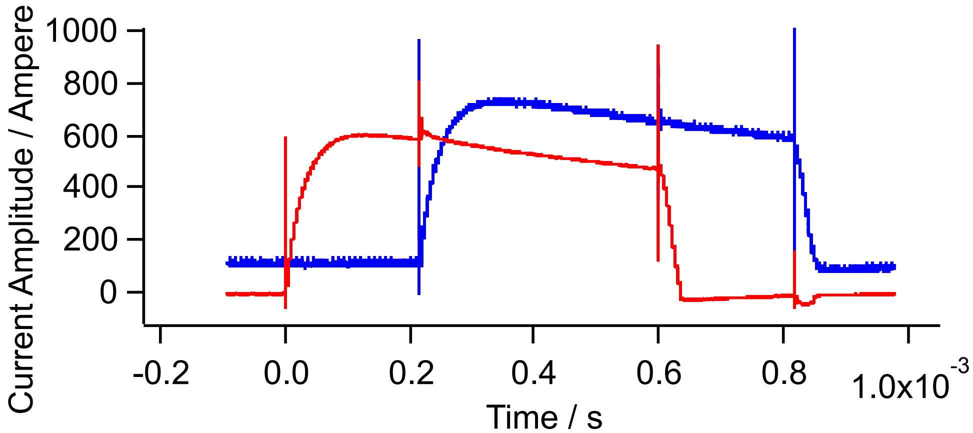
Trapping of CH_3

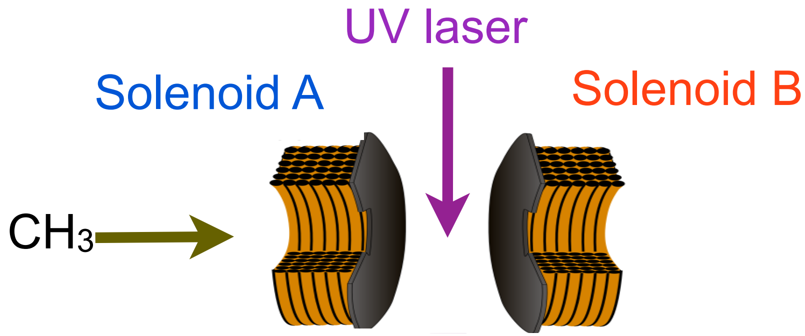


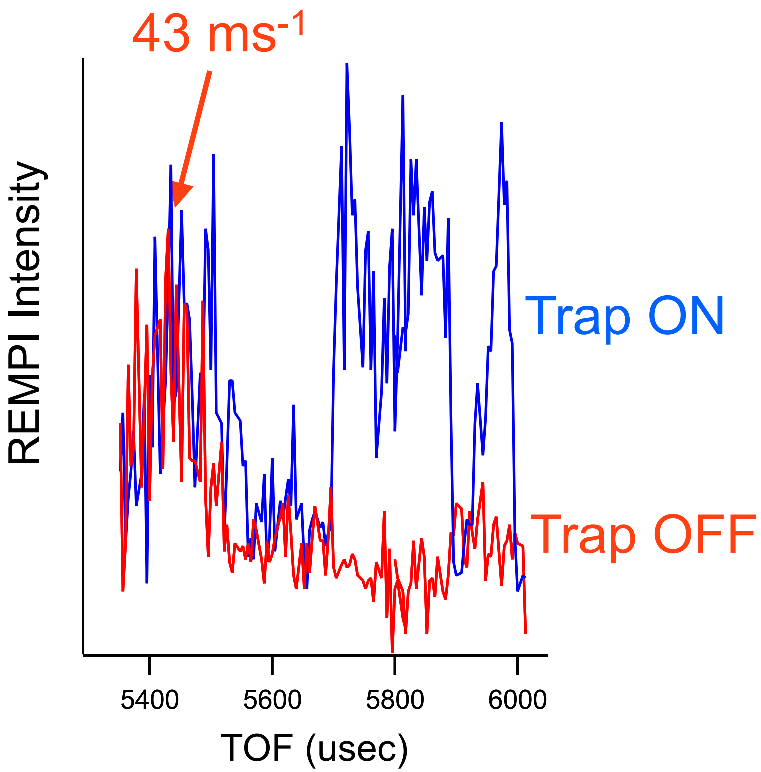




Trapping of CH_3







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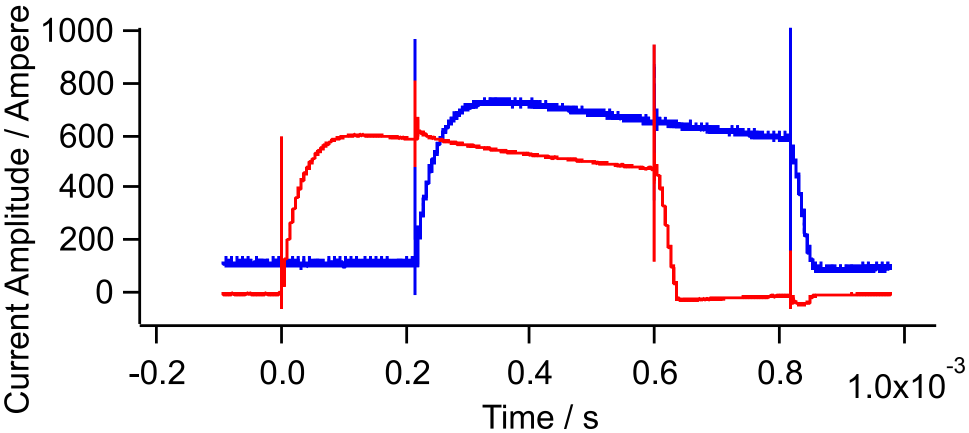

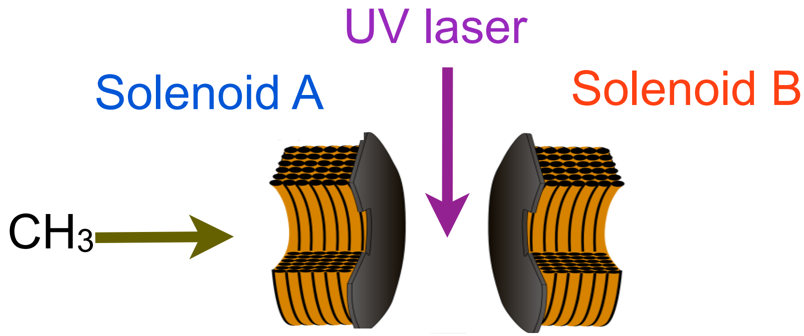

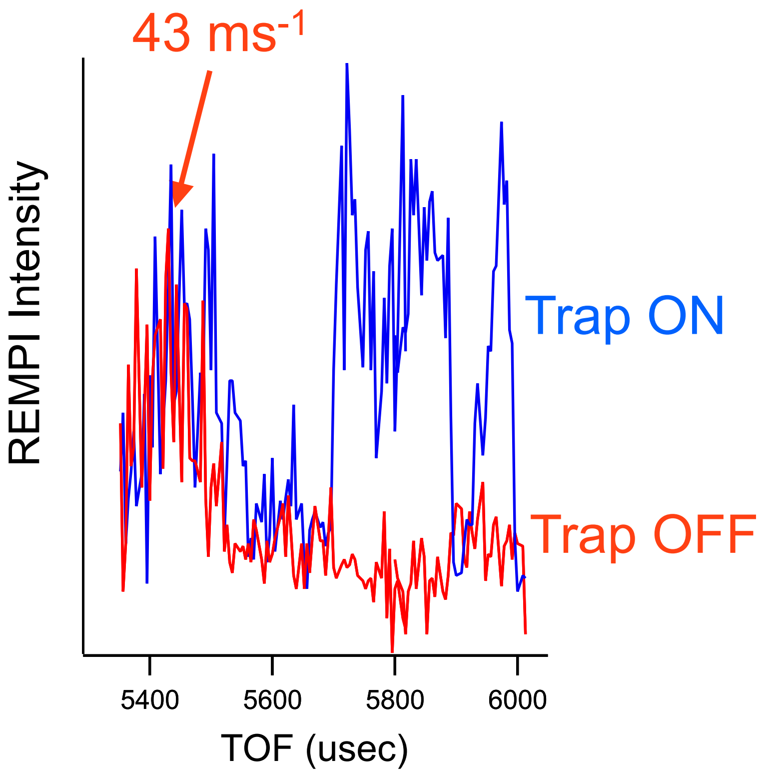

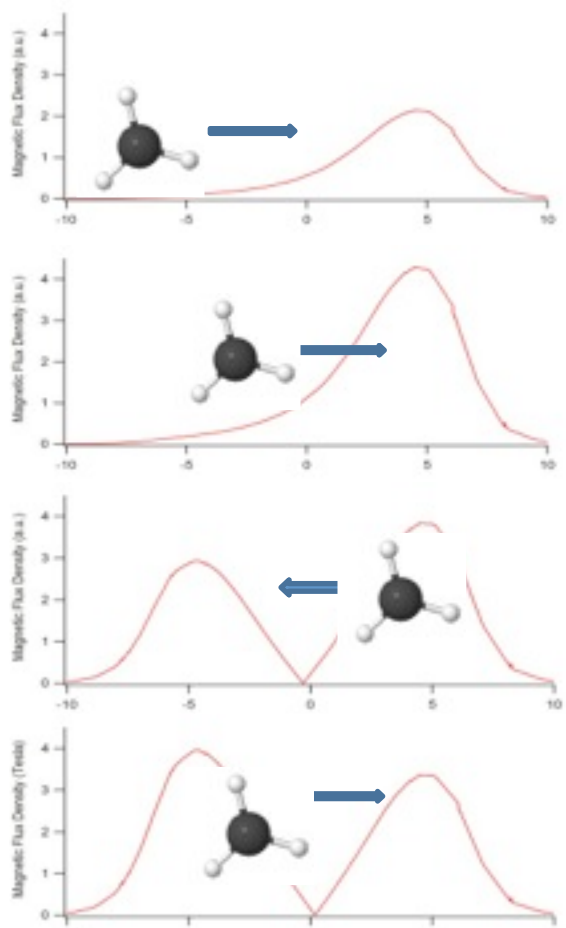

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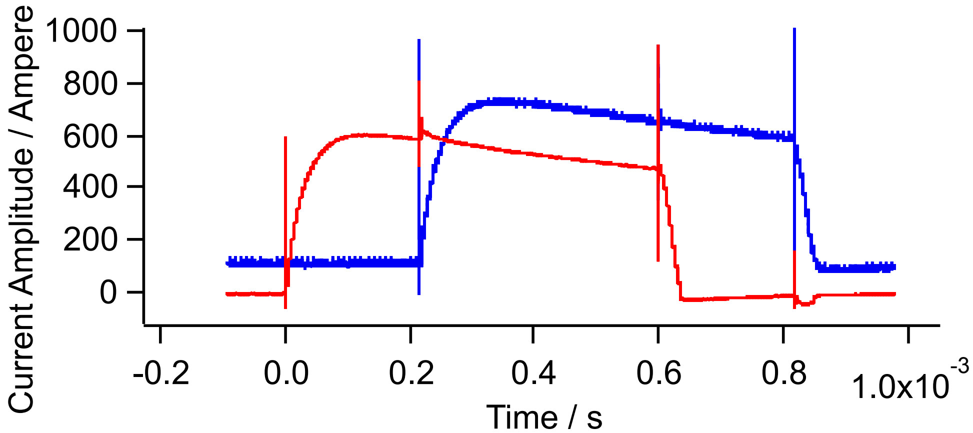

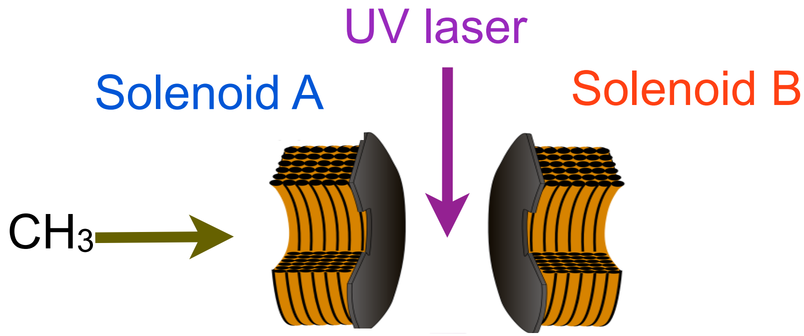

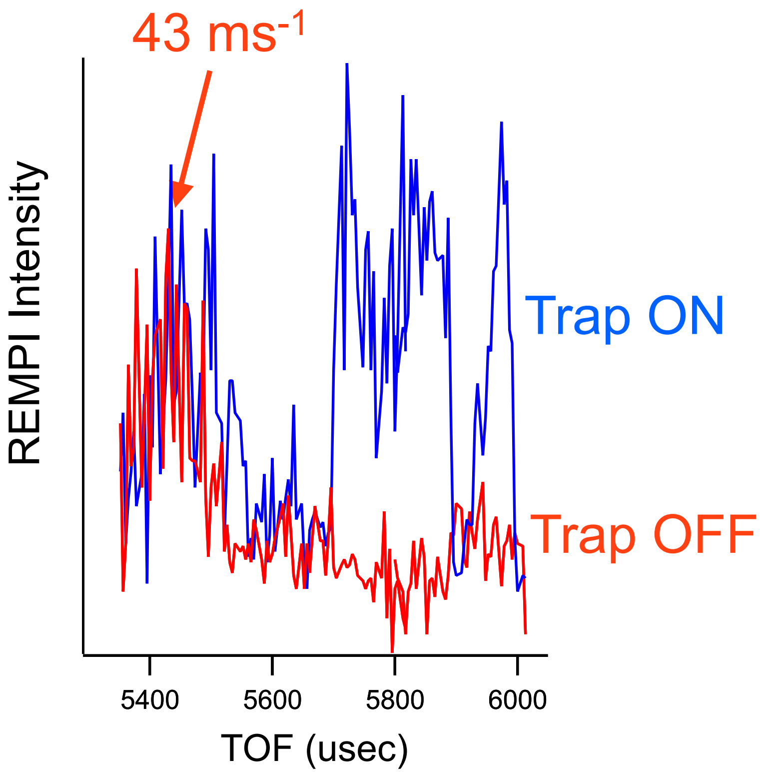

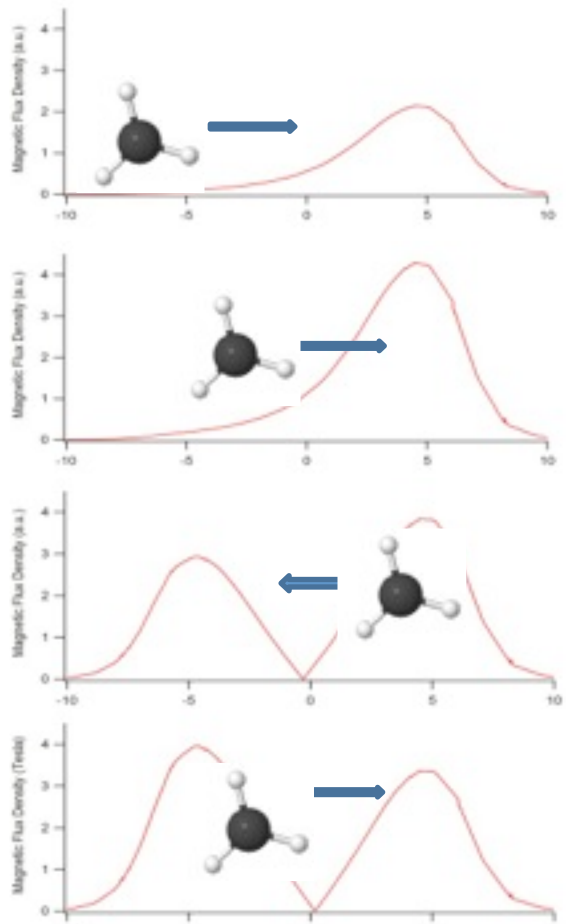

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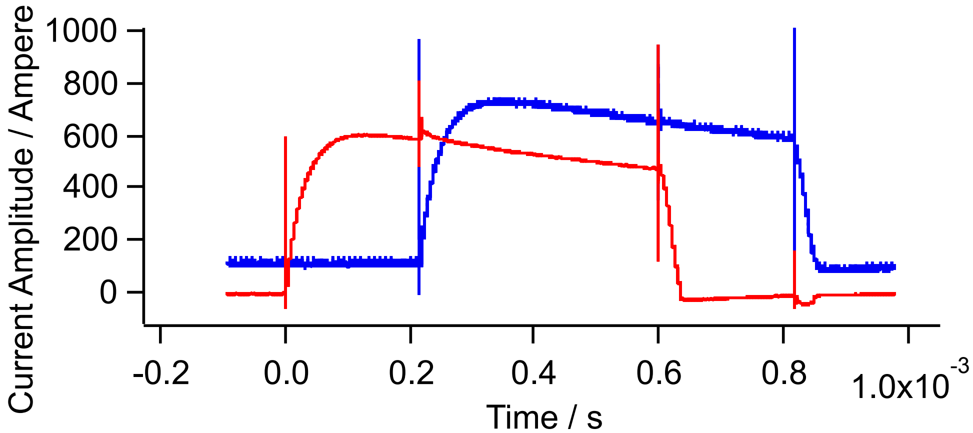

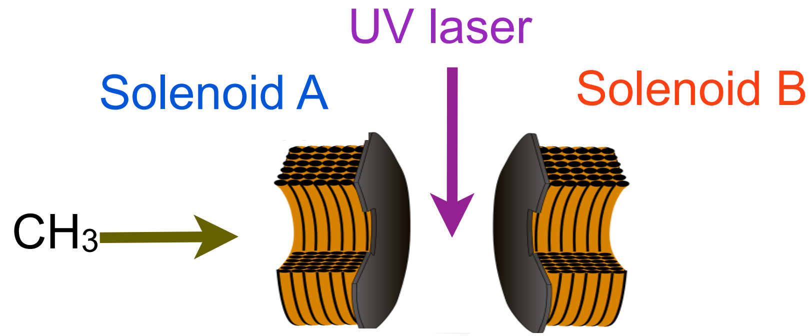

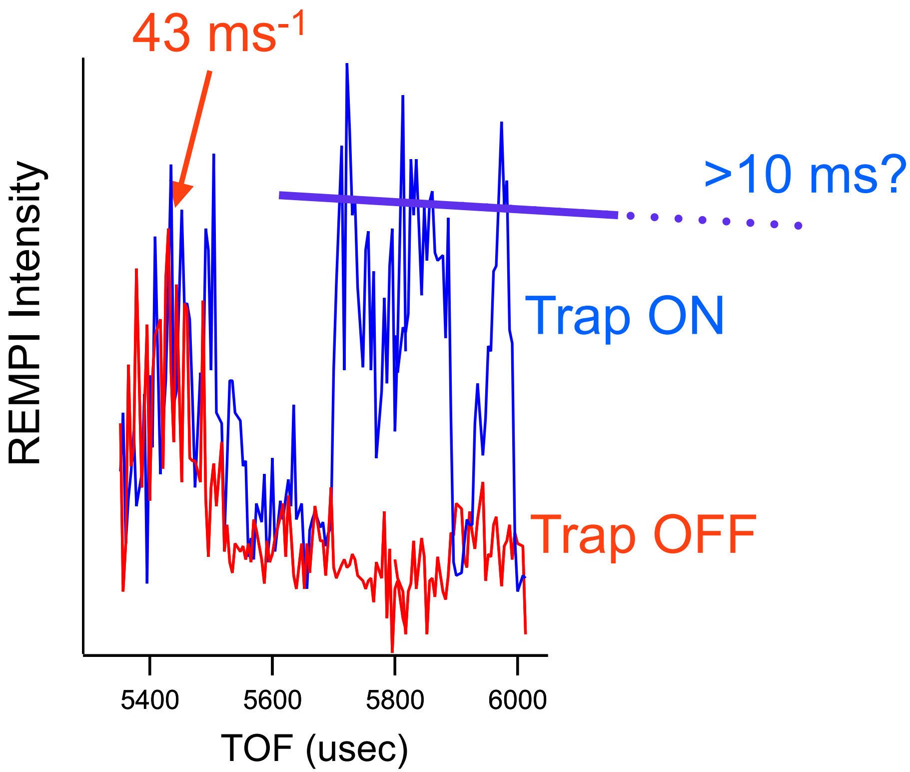

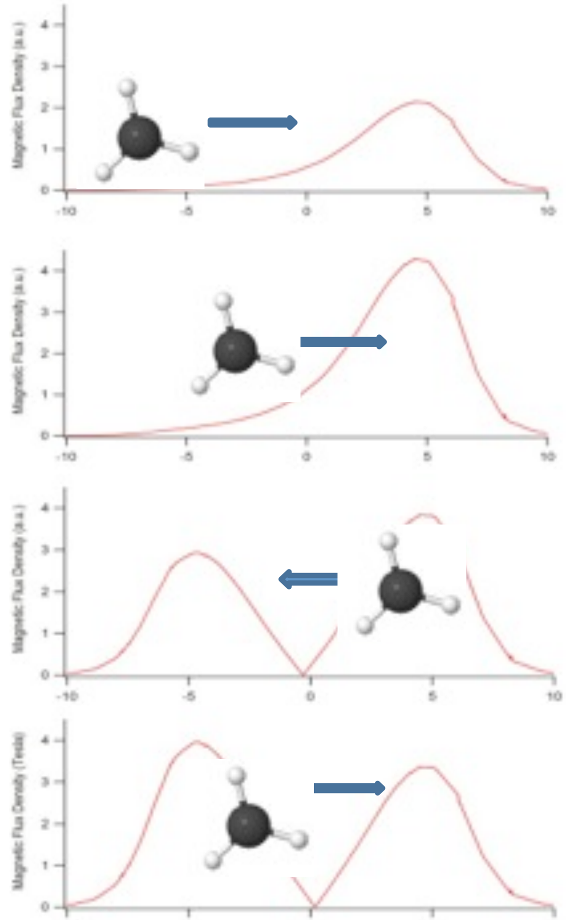

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CH_3 radicals are now spatially confined !

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$v_{\parallel} =$ **40 ms^{-1}** $v_{\perp} \sim$ **1 ms^{-1}**

volume 4 mm^3

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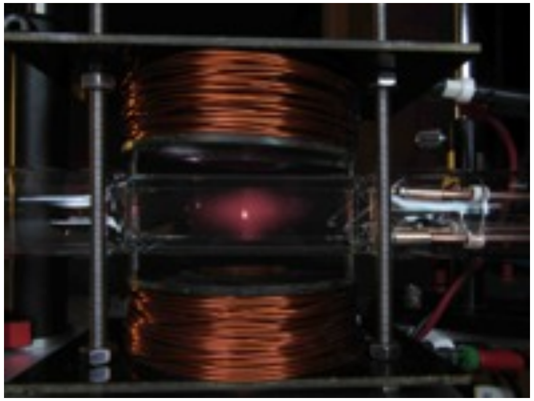

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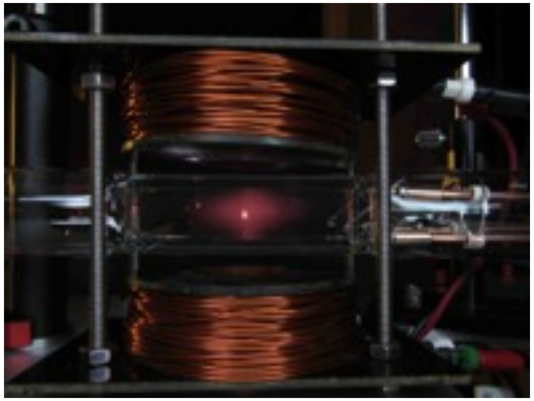

Any non-singlet state of molecules can be decelerated and trapped.

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